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SBRG

STUDY RELATING TO THE PRODUCTION OF OIL FROM LOW RANK CANADIAN COALS BY DIRECT LIQUEFACTION - PHASE IV

FINAL REPORT

~~CO~~NFIDENTIAL

BY

SANDWELL BEAK RESEARCH GROUP

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SBRG

23 June 1983

Dr. J. Kelly, Ph.D.
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Dear Jim:

We are pleased to submit to you the original and four (4) copies of the final report on The Study of the Production of Oil from Low Rank Canadian Coals by Continuous Liquefaction, Phase IV. The report contains a complete summary of work and results obtained during the period covered. We included in it your comments on the draft report presented to you in May.

Under the terms of this contract, Her Majesty agrees to keep confidential the material contained in this report for 24 months.

We hope this satisfies your requirements. Upon your acceptance of this report, DSS will proceed with outstanding payments to us.

Yours very truly

SANDWELL BEAK RESEARCH GROUP

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80 p.p.

PREFACE

This report covers the work conducted by SBRG under the shared-cost coal conversion program of CANMET during the period from April 16, 1982 until March 31, 1983.

This work program was a continuation of prior work conducted by SBRG using low rank Canadian coals. The program included the operation of a 0.1 TPD continuous direct liquefaction process development unit using Estevan lignite. The main activities and achievements reported herein include:

1. design and construction of the first generation prototype equipment to operate in a batch mode to demonstrate the process concepts of the Sandwell product recovery and solids separation process,

2. the new process module was successfully tested in an integrated mode of operation with the PDU,

3. the Sandwell process concepts were successfully demonstrated within the constraints of the equipment,

4. comparative runs were conducted using both hydrogen and syngas reducing atmosphere.

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1.0 INTRODUCTION

At the conclusion of the Phase III of the Coal Conversion Program 500 hours of PDU operation had been completed using 4 Canadian coals under 18 sets of operating conditions. Operation of the continuous system under coal derived solvent recycle was achieved with very encouraging results. These conditions had resulted in better overall conversion and liquid products yield than was found in the once through studies. In addition, on 27 February 1982 Sandwell filed for a provisional British patent for a novel process for the separation of solids from primary liquefaction products. The successful development of the program in these previous years motivated continuation of work along the same objectives, in order to advance the development work on the novel process for solids separation, improved characterization of the product and solvent quality as well as further investigation of use of carbon monoxide/ hydrogen mixture as reducing agent. Three continuous runs were conducted in solvent recycle mode using coal derived product as start up solvent.

The first of these runs was carried out at 440°C and 24.1 MPa using vacuum distillation for recovery of a solvent fraction for 63 hours, accomplishing 6 passes through the unit. Part of the final product was then distilled and the 250-450°C fraction was hydrotreated catalytically, in once through mode using HDS 1442 catalyst at 400°C and 24.1 MPa. The objective was to improve the hydrogen content of the product to be used in the subsequent runs.

A second recycle run was undertaken using a mixture of hydrogen and carbon monoxide at similar conditions. This run could not be completed due to mechanical problems. The results obtained in the first period of the run indicate a very high coal conversion and liquid yield, and warrant further investigation in the continuation of the program.

A process simulation of the solids separation process was carried out in the third run using first generation prototype equipment. The simulation represents one of the possible process configurations described in the patent application for the solids separation process. Full simulation of the process concepts would require special equipment not available at this stage of the program. The equipment which was used was designed and constructed within the financial restrictions of the program and provide an initial demonstration of some of the fundamental process concepts.

Solids separation was integrated with the operation of the liquefaction PDU to provide a complete self sustaining coal liquefaction plant. The solids separation process was successfull in removing solids, asphaltenes and pre-asphaltenes. At the same time it separated sufficient recycle solvent for the slurrying of coal to maintain solvent balance.

The asphaltenes and pre-asphaltenes were separated in the form of a concentrated stream of these products from which the bulk of the solids (THF insoluble material) had been removed. This product stream can either be utilized directly, as for instance a solid or liquid fuel for power generation, or it may be upgraded to distillates. The unconverted coal and ash (solids) were in essence completely separated from the coal derived products and coal derived recycle solvents. The final solids by-product carries with it only very small amounts of liquid product. This results in near maximum liquefied coal material recovery, which coupled with a high total conversion and low gas production in the liquefaction reactor, provides for very high yields.

2.0 SUMMARY

The direct liquefaction of coal promises to be an efficient and economic process for the production of liquid fuels. In Canada, large resources of reasonably priced low rank coals could supply the necessary feedstocks to a synfuels industry. To realize the potential of those resources, it is necessary to develop a technology which can efficiently transform these coals into quality liquid fuels.

The three previous years work in this program, dedicated to developing a technology for these coals, had resulted in the completion of a 0.1 TPD continuous liquefaction process development unit which is an essential tool for advanced process development. With this facility, an ambitious research program was undertaken on Canadian lignite and sub-bituminous coals. The following describes the results of Phase IV in terms of the operation of an upgraded PDU, research on the primary liquefaction stage of the process, demonstration of the Sandwell solvent recovery and solids separation process and a review of the economics of the direct liquefaction of low rank coals.

SOMMAIRE

La liquéfaction directe du charbon promet d'être un procédée économique et efficace pour la production des combustibles liquides. Le Canada a des grandes quantités de charbons pauvres à un prix raisonable qui peuvent fournir les matières premières pour une industrie de combustibles liquides. Afin d'exploiter ce potentiel il faut développer une technologie qui pourra transformer de façon efficace ces charbons en combustibles liquides de bonne qualité.

Les trois premières années de ce programme visant le développement d'une technologie pour la valorisation des charbons pauvres ont eu comme résultat la construction d'une installation pilote de 0.1 tonne par jour; ce qui est un outil essentiel au développement poussé d'un tel procédé. GRSB s'est engagé dans un program de recherche très ambitieux utilisant cette unité pour la liquéfaction des lignites et des charbons sub-bitumineux de l'ouest du Canada. Dans les pages suivantes sont presentés les résultats de la quatrième étape de ce programme en ce qui concerne l'opération de l'unité pilote, les améliorations portées à l'équipement, des recherches sur la première étape de la liquéfaction, la démonstration du procédé Sandwell pour la récupération du solvent et la séparation de matières solides dans le produit et aussi une analyse économique de la liquéfaction directe des charbons pauvres.

2.1 PDU OPERATION

During the second year of operation of the PDU, the unit operated for 120 hours and generally performed very well. Numerous improvements were made throughout the year to improve reliability, safety, sampling, flow measurement and control, operational range and efficiency of operation.

OPERATION DE L'INSTALLATION PILOTE

Durant la deuxième année de l'opération du pilote, l'unité fonctionna pendant 120 heures et son rendemment fût trés satisfaisant. De nombreuses modifications ont été portées à l'équipement pour améliorer la sécurité au travail, l'échantillonage, la mesure et le contrôle des débits, l'étendue des conditions d'opération et l'efficacité des opérations.

2.2 LIQUEFACTION RESULTS

The PDU was operated in Phase IV of the program to further investigate the liquefaction properties of the subject coal under product recycle conditions. For comparison purposes the same coal -Estevan Lignite- was utilized as the reference coal. The same operating conditions used in Run 10 as reported previously were repeated. Run 512 duplicated the conditions of Run 10 in all respects including product recycle by vacuum distillation. Run 514, employing the same reaction conditions as Runs 10 and 512, used for the first time a simulation of the Sandwell product recovery solids separation process. Finally, Run 513, investigated the use of syngas (CO and H_2 mixture) in lieu of pure hydrogen. The following table summarizes the main findings.

Tests with Hydrogen

The two runs conducted in Phase IV gave very similar results in terms of overall conversions 90.3 and 90.0% and total gross liquid yields 63.8 and 67.1%. Generally however, these were about 5% lower than the results of Run 10 conducted approximately one year earlier on the same batch of lignite. Since all other operating conditions were roughly equivalent, the lower conversions and yields are attributed to further aging of the lignite, although it had been stored under an inert gas environment.

The tests confirmed the benefit of recycle of coal derived liquids as solvent material giving better conversion and yields. Regardless of the separation system employed

RESULTATS

Dans cette quatrième étape, l'unité pilote a été utilisée afin de poursuivre l'investigation de la liquéfaction du charbon avec recyclage du solvent. Pour faciliter la comparaison des résultats, le même charbon -le lignite d'Estevan- a été utilisé comme charbon de référence.

L'essai No 512 fut effectué dans les mêmes conditions que l'essai No 10 qui a été decrit dans le rapport pour l'année dernière. Le même mode de récupération du solvent, c'est à dire, la distillation sous vide, a été employé. L'essai No 514 effectua les mêmes conditions d'opération pour l'étape de liquéfaction, mais pour la première fois une simulation du procédé Sandwell de séparation des solides fut utilisée pour la récupération du solvent.

Finalement, l'essai No 513 a eu comme but d'étudier l'effet du gaz de synthèse au lieu de l'hydrogène pure. Le tableau suivant présente les résultats principales

Les essais avec l'hydrogéne

Les deux essais utilisant l'hydrogène pur comme agent d' hydrogénation dans la quatrième étape ont donné des resultats comparables en ce qui concerne la conversion du charbon: 90.3 et 90.0%; et le rendement en liquide: 63.8 et 67.1%. Ces résultats sont environ 5% de moins que les résultats obtenus dans l'essai No 10 qui a été effectué une année auparavant avec le lignite du même lot. Quand on considère que les conditions d'opération furent les mêmes, ces résultats plus bas doivent être attribués aux vieillissement du charbon même si il était entreposé sous atmosphère inerte.

the recycle solvent was a roughly equal composite of primary reactor product (oils, preasphaltenes, asphaltene, ash and unreacted coal) and a middle to heavy distillate cut from the primary products. Although six passes were completed, the test periods were of insufficient duration to establish steady-state conditions. In the recycle solvents the concentration of non-distillates were increasing progressively although at slower rates. However, extended tests are required to determine the preferred combination and selection of recycle solvents to product stable operation over long periods.

Clearly the recycle of non-distillates, unreacted coal and ash is beneficial but the limit needs to be determined by additional solvent studies and multiple pass operation.

Synthesis Gas Mixtures

A preliminary once-through study with syngas during the previous years program gave inconclusive results. Furthermore, it had been decided to concentrate first on operations with hydrogen in order to establish a strong basis of comparison for eventual experimental work using CO and H_2 mixtures. Previous studies by SBRG and others have demonstrated the potential advantage of syngas mixtures particularly for highly oxygenated coals.

Although operating problems prevented a full recycle experiment, the results of Run 513 using a mixture of CO/H_2 in a feed gas ratio 40/60 resulted in superior liquid yields compared to using hydrogen only. Despite slightly lower conversion 88.2% compared to 90.3%,

Les essais ont confirmé l'effet bénéfique du recyclage de produits provenant du charbon dans le solvent.

Indépendamment du mode de séparation utilisé le solvent recyclé était un mélange à peu près égales de produits du réacteur comme tel (incluant des huiles, asphaltenes, pré-asphaltenes, cendres et le charbon non convertit) et une fraction de distillation correspondant à une huile lourde. En dépit du fait que six passes ont été complétées, les réactions n'ont pas atteint un régime de stabilité (steady-state conditions). La concentration de produit non-distillable dans le solvent recyclé a augmenté lentement. Des essais de longue durée sont requis si on veut déterminer la meilleure combinaison et sélection de solvents pour arriver par recirculation à un régime de stabilité.

C'est évident que le recyclage des produits non-distillables, du charbon nonconvertit et des cendres a un effet bénéfique sur la liquéfaction, mais la limite supérieure du contenu de ces produits dans le solvent doit être déterminée par des études approfondies sur la qualité du solvent et sur l'opération prolongée de l'unité.

Les essais utilisant le gaz de synthèse

Une étude préliminaire utilisant un mélange de CO et H_2 sans recyclage du solvent avait été effectuée dans le programme précédent, sans avoir donné des résultats concluants. A ce temps là il avait été decidé d' étudier d'abord le procédé utilisant de l'hydrogène pur afin d' établir une base de comparaison avant de s'occuper de l'aspect gaz de synthèse.

Des études préliminaires effectuées par GRSB dans une autoclave ont indiqué l'avantage du gaz de synthèse au lieu de l'hydrogène pur, spécialement avec les charbons à haute tenure en oxygène . L'essais No 513 a été effectué sur l'unité en continu, avec un mélange 60/40 d'hydrogène et monoxide de carbone.

TABLE 2.1: COMPARISON OF RESULTS FOR RECYCLE RUNS NUMBER[1], 512, 513 and 514

RUN NUMBER	10	512	513	514
Experiment No	17	519/1	522	523
Preheater Temperature (°C)				
Slurry	300	300	290	300
Gas	350	350	300	350
Reactor Temperature (°C)	440	440	440	440
Reactor Pressure (MPa)	24.2	24.2	24.2	24.2
LHSV (hr^{-1})	0.98	1.08	0.94	0.91
MAF Coal flow rate (kg/hr)	1.61	1.47	1.48	1.27
Solvent/Coal (wt ratio)	2.0	2.1	1.6	2.3
Gas/ Liquid ratio (L/kg)	192	174	168	181
Mole % H_2 in feed gas	97	100	59	100
Conversion (% wt MAF coal)	95.5	90.3	88.2	90.4
Yields (% wt MAF coal):				
Liquid (gross)	68.0	63.8	72.4	67.1
C_1-C_3	5.2	8.5	7.3	6.4
H_2O	13.7	12.9	(16.8)	10.0
$CO + CO_2$	10.2	9.6	4.7	10.1
H_2S	0.4	0.2	0.3	0.2
H_2 consumption	(1.9)	(4.7)	(1.9)	(3.9)

% MAF Coal recovered

1) Reference run conducted in 1982 (Phase III of Project). Data for this run are averaged over 5 passes.

the liquid yield was 72.4% almost 10% higher than obtained in the runs with pure hydrogen. Furthermore, the distillate yields as measured by the hexane soluble fraction of the product were substantially improved as a result of using syngas mixtures. The products appear to be lower in heteroatoms as a result of syngas presence. The benefits observed are considered to be related to the presence of nascent hydrogen produced as a result of the water shift reaction as well as of the scavenging reactions of carbon monoxide with oxygen ether bonds present in the coal, leading to deoxygenation.

The results strongly suggest that further studies with syngas be conducted under recycle conditions and extended runs.

2.3 ADVANCES IN SOLIDS SEPARATION TECHNOLOGY

Run 514 was conducted using a first generation prototype equipment designed to provide an initial demonstration of some of the fundamental process concepts of the Sandwell advanced separation process. The objective of the process is the separation of solids (ash and unreacted coal) leaving both the distillate and non-distillate liquid products relatively solids free. Through the use of vacuum distillation, solids free distillates are obtained while the nondistillable liquid products remain together with the solids in the residue, product recoveries are generally in the order of 50%.

Run 514 confirmed the fundamental concents of the Sandwell process. For three passes, the amount of primary product liquid recovered exceeded 98% thus proving the efficiency of the process as a superior means of product recovery.

L'essai ne fût pas completé à cause de problemes mécaniques mais les résultats obtenus pendant le temps d'opération furent nettement supérieure aux essais avec l'hydrogène pur. La conversion du charbon calculé à base de fraction soluble en tetrahydrofurane était 87.5%, et le rendement en produit liquide était 72.4%, ce qui est environ 10% de plus élevé que les rendements en produit liquide obtenus avec l'hydrogène pur. En plus, le rendement en produit distillable mesuré par la fraction du produit soluble dans l'hexane a sensiblement augmenté avec l'utilization du gaz de synthèse.

Les produits obtenus semblent avoir moins d' hétéroatomes quand le gaz de synthèse est utilisé. Il est possible que tout ces bénéfices soient reliés à la présence d'hydrogène nascent produit par la reaction de "shift" et aussi aux réactions de balayage des liens éther-oxygène du charbon par le monoxide de carbone.

Ces resultats nous suggèrent fortement de poursuivre ces études avec le gaz de synthèse en faisant des essais de plus grande durée et avec recyclage.

PROGRES EN TECHNOLOGIE DE SEPARATION DES SOLIDES

Une opération prototype de première génération fut concue pour démontrer les idées principales du procédé Sandwell pour la séparation des solides. Ce procédé vise la séparation des cendres et du charbon non-converti donnant un bon rendemment en produit liquide avec presque pas de solides. Par contraste, la distillation sous vide effectue une séparation sur le distillat seulement, laissant environ 50% des liquides avec les residues de distillation dans un état non-séparé.

L'essai No 514 confirma les idées fondamentales du procédé Sandwell. Plus de 98% des liquides furent séparés et récupérés, démontrant ainsi l'efficacité de ce procédé pour la récuperation des produits de liquéfaction.

2.4 ECONOMIC ANALYSIS

An economic model was developed to provide a relative comparison between the cost of a commercial plant based on vacuum distillation for product recovery and separation by the Sandwell process. By recovering more liquid product suitable for further upgrading to distillate fuels, the model estimates a 28% reduction in the cost of producing liquid fuels as a result of the Sandwell process.

ANALYSE ECONOMIQUE

Les coûts d'une unité commerciale utilisant la distillation sous vide comme méthode de séparation fûrent comparés au coûts d'une unité utilisant le procédé Sandwell. Puisque le procédé Sandwell permet un rendement plus important en produit liquide susceptible au raffinage l'analyse promet une réduction de 28% dans les coûts de production si l'on utilise le procédé Sandwell.

3.0 PDU UPGRADING

3.1 PERFORMANCE REVIEW AND MODIFICATIONS

The monitoring of the equipment performance and the increasing demands placed on the PDU, led to a number of modifications. As a result the system was provided with increased safety, flexibility and reliability in extended operation, and improved material balance through better gas measurement and analysis.

The following is a list of the changes and additions to the equipment, completed during the period of the report.

A flow diagram of the PDU updated to include the modifications is shown in Figure 3.1.

Installation of New High Pressure, High Temperature Receiver

A review of the design and the operation of this step revealed limitations in the allowable working conditions for the hot high pressure receiver. The old receiver was replaced by a new vessel designed for operation at 450°C with an increased safety factor.

The new vessel was custom fabricated (body made of 316 stainless steel sch. 160 and cap made of alloy steel 4140 HR. HT - to 4193, Grade B7) equipped with compression closures. After being commissioned in Run 512, the new receiver was operated at a higher liquid (bottom) temperature (240-260°C) in order to check its performance with regard to light ends and water flashing. The slurry inlet was extended 15 inches into the vessel, and a thermocouple well was provided in the bottom for the liquid temperature monitoring. The level control is maintained using the same gamma-ray indicator installed for the old vessel.

Automatic Pressure Let-down for Light Product

In the past, the collection of the light product was done by maneuvering manual valves in such a way that a 2 liter vessel (M3X3) which was periodically opened to pressure for a few minutes until all the light product collected in the cold receiver (M3X2) flowed down. Then this vessel was isolated from the rest of the unit and vented carefully until the pressure descended to ambient. When this maneuver was complete, the light product was emptied in the receiver.

In order to increase safety of operation and also to mitigate the pressure unbalancing due to the need to pressurize a 2 liter vessel every time the light product was collected (hourly) the following changes were made:

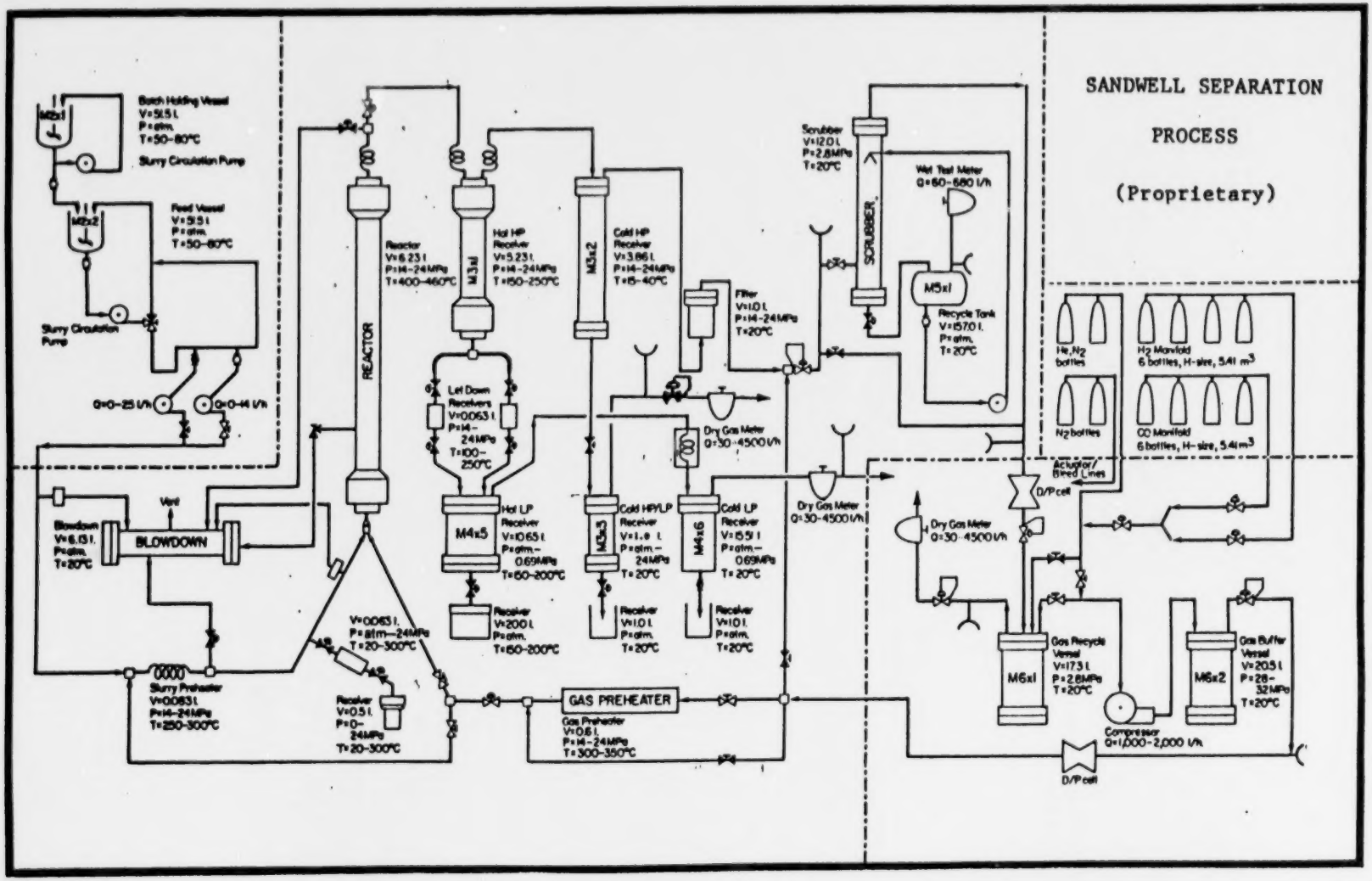


Figure 3.1 **SBRG Continuous Unit Flow Diagram: Volumes and Typical Operating Conditions**

- the 2 liter M3X3 was repaced by a 1 liter vessel
- a system of interlocked actuated valves was installed in addition to the manual ones and their operation was designed to prevent pressure loss in the unit by faulty operation of let down.
- a pressure regulator was installed in the vent lines to protect the gas meter from pressure surges.

Additional Instrumentation

A new pressure transducer was installed in the slurry line downstream of the slurry preheater, before the reactor inlet to replace the PPI Astragauge which failed repeatedly in the past.

A separate alarm panel was designed and added to the main control panel.

In order to prevent large flow variations of gas feed a low flow alarm was added.

3.2 NEW EQUIPMENT RELATED TO SOLVENT RECYCLE OPERATION

A large slurry preparation vessel (100L, height/diameter ratio 2/1) was installed on the upper deck for solvent and slurry mixing.

The research area was extended by a 15' x 15' room to house the solvent recovery module. The vacuum distillation unit was moved into this new room. A new control panel was built and utilities and power, vacuum, compressed air, nitrogen purge, cold and hot water and vent lines were piped in.

A 100L, medium pressure vessel provided with electric heating, mixing and nitrogen purge. The vessel was custom fabricated of 316 stainless steel.

The existing centrifuge was provided with a variable speed motor, new brake system and electric heating.

A vacuum evaporator ($0.5M_2$) provided with electric heating, condensor and glass collecting vessels was designed, fabricated and it was connected to the existing vacuum pump.

All new equipment was commissioned in March 1983 and it performed satisfactory.

Additional Capability for Gas Chromatographic Analysis

A new Perkin Elmers Sigma 3B Gas Chromatograph was added to the analytical capabilities dedicated to the PDU operation. The new GC will allow determination of ammonia, hydrogen sulfide and C_4 hydrocarbons. The commissioning and calibration of the apparatus were complete before the three runs were conducted.

4.0 BATCH TESTING

4.1 EQUIPMENT MODIFICATION

The existing hot charge reactor was modified in order to alleviate slurry charge and gas leakage control problems. The modifications included installation of a calibrated hydraulic piston for slurry charging, redesign of the main reactor lid, and addition of a new custom designed cold receiver for the reaction product. The new receiver has a screw cap, easy to take apart and reassemble for removal of thick, sticky or solidified product.

4.2 TESTING PROGRAM

This year's testing program had to be limited, due to lack of time and only six tests were carried out as part of a systematic program for determining correlations of batch results to the continuous system performance. This program will be continued and will include testing at a range of conditions, using a variety of solvents (such as anthracene oil, tetraline, coal derived recycle fractions) and a variety of coals.

The final goal of the program is to establish a data base allowing us to use the batch reactor to forecast potential of coals for direct liquefaction in continuous system.

Four tests were undertaken using the modified reactor with Estevan and Willow Bunch lignites and using anthracene oil as solvent. In addition, two tests were carried out to investigate the effect of "red mud" (an iron rich waste stream from alumina extraction) as catalyst for hydrogenation of a heavy product fraction obtained on the continuous system.

The compositions of the two coals are presented in Table 4.1. The Estevan lignite used was part of the shipment used for the continuous testing, the sample of Willow Bunch coal was supplied by Dr. R. Ranganathan of Saskoil. The latter sample was ground before testing. The anthracene oil used as solvent was part of the shipment used for the continuous tests.

The reaction conditions and some results obtained in the tests are presented in Table 4.2.

TABLE 4.1: ANALYSIS OF COALS AND SOLVENTS USED IN BATCH TESTING

COAL	Estevan Lignite	Willow Bunch Lignite
Proximate Analysis (% wt)		
Moisture	21.8	35.9
Volatile (mf)	39.9	30.5
Fixed carbon (mf)	43.1	32.6
Ash (mf)	17.0	36.9
Ultimate Analysis (% wt maf)		
Carbon	71.4	69.8
Hydrogen	4.9	4.8
Nitrogen	2.2	0.8
Sulphur	1.3	1.6
Oxygen (by diff)	20.2	23.0

SOLVENT	Anthracene Oil
Distillation Fraction, % wt	
(IBP) 135-200°C	3.5
200-270°C	12.0
270-300°C	7.0
300-350°C	40.9
350-400°C	26.6
400-412°C	4.5
412°C	5.5
Ultimate Analysis, %	
C	91.1
H	5.7
N	1.0
S	0.6
O (by difference)	1.6
O (by neutron-activation)	2.0

TABLE 4.2: BATCH TESTING REACTION CONDITIONS AND RESULTS

Test No.	10	11	14	15	19	20
Solvent	Heavy product containing ash and unreacted coal		AO	AO	AO	AO
Coal Type			Willow Bunch		Estevan	Estevan
Temperature, °C	430	425	425	400	425	425
Pressure, MPa	22.0	20.7	23.1	24.8	21.1	24.1
Reaction time, min.	45	45	45	25	60	60
Catalyst	-	Red Mud	-	-	-	-
Conversion	46	40.2	90.7	71.6	83.7	84.8
Yields, % maf coal						
liquid	19.4	28.1	39.7	40.7	49.4	46.4
H_2O			13.6	11.9	18.1	19.1
CO, CO_2	0.9	0.8	18.2	15.5	10.9	12.7
C_1-C_3	33.0	21.9	20.9	5.8	8.7	9.3
H_2	(7.3)	(10.6)	(1.7)	(1.8)	(3.4)	(3.5)

4.3 DISCUSSION OF THE RESULTS

The testing of Willow Bunch and Estevan lignite at similar conditions indicate a higher reactivity for the Willow Bunch (90.7% conversion in 45 minutes compared to only 84% in 60 minutes for the Estevan). The liquid yields obtained from Willow Bunch lignite though, were relatively low in favour of high gas yields. The low liquid yield compounded with the very high ash content of the initial coal will be serious arguments limiting the potential of Willow Bunch lignite as feedstock for direct liquefaction.

The results obtained with the Estevan lignite in the batch mode compared to the once through continuous testing carried out at similar reaction parameters on the PDU, reproduced well the level of conversion, but resulted in lower liquid yield and higher deoxygenation products yields as shown in Table 4.3.

The two tests (No. 10 & 11) in which no fresh coal was added and only heavy (M4X5) product including ash and unconverted coal obtained in Run 11 on the PDU after eight passes through the unit, confirmed the preliminary results obtained in continuous mode, namely that the organic insolubles recycled continue to be converted, albeit to a lower degree than in the initial pass, with high hydrogen consumptions and high gas yields. An interesting observation, which also agreed with the continuous tests is that the presence of the Red Mud enhances the conversion of preasphaltenes into asphaltenes and further into oils as shown in Table 4.4.

TABLE 4.3: COMPARISON OF YIELDS FOR BATCH AND CONTINUOUS TESTING

TEST	BATCH MODE		CONTINUOUS MODE		
			ONCE THROUGH	ONCE THROUGH	RECYCLE
	No. 19	No. 20	No. 3	No. 4	No. 17
Conversion	83.7	84.8	84.5	84.3	95.5
Yields, % maf coal					
Liquid	49.4	46.4	64.2	58.8	68.0
H_2O	18.1	19.1	10.7	12.2	13.7
CO, CO_2	10.9	12.7	5.7	9.9	10.2
C_1-C_3	8.7	9.3	4.6	4.6	5.2
H_2	(3.4)	(3.5)	(1.3)	(1.2)	(1.9)

TABLE 4.4: HYDROCRACKING OF LIGNITE DERIVED LIQUIDS USING RED MUD - RESULTS

Exp. No.	10	11	18-08	18-09
Mode	← batch →		← continuous →	
Catalyst added	none	red mud	none	red mud
conversion	49.6	41.0	37.5	20.5
H_2 consumed	7.3	10.3	3.6	3.1
Yields % maf:				
total liquid	24.7	28.0	12.1	18.8
H2O	n.d.	n.d.	6.3	2.3
CO + CO_2	0.9	0.8	12.2	1.5
C_1-C_3	31.5	23.0	9.9	7.1
hexane soluble	17.3	27.2	17.7	24.2
asphaltene	7.8	13.2	(3.0)	2.4
preasphaltene	(0.4)	(12.8)	(2.5)	(7.8)

5.0 PRODUCT CHARACTERIZATION

In search of more efficient methods of solvent quality monitoring two methods were studied during the last year.

5.1 SIMULATED DISTILLATION

Simulated distillation has been widely used as a method for obtaining boiling point data of petroleum fractions. Although recent references exist in the literature for the application of simulated distillation to coal-derived liquids, there is still controversy regarding the applicability of this technique, especially for oxygen containing compounds. A number of laboratories we contacted have been using simulated distillation strictly to monitor product stability.

SBRG started work on simulated distillation for the purpose of following solvent quality for the primary liquefaction step as well as for the solids separation process. This section outlines the work done to determine the applicability of this technique to characterization of our coal derived products.

Our simulated distillation system consists of a Perkin Elmer Sigma 3 gas chromatograph with dual FID detectors and a Sigma 10 data acquisition console. The columns used were 46 cm long, 0.32 cm diameter, stainless steel, packed with 10% UCW-98 on Chromosorb W A/W 80/100. Operating conditions were as follows:

column temperature	- initial	: 70°C
	- final	: 350°C
	- rate	: 8°C/min
detector temperature		: 375°C
injector temperature		: 375°C
carrier gas		: N_2
sample size		: 0.2 - 1.0 μl
analysis time		: $\sim$35 minutes

Calibration was accomplished with a mixture of n-paraffins from C_7 to C_{40} covering the boiling range 98°C to 525°C. A calibration curve showing retention time versus atmospheric boiling point is presented in Figure 5.1. The curve is linear except for a considerable curvature at the lower boiling points due to the high starting temperature (70°C). Since we are interested in product boiling above 205°C no effort was made at this time to start at a lower temperature. Retention times of certain ring type compounds versus equivalent atmospheric boiling points were also obtained and are included on the calibration curve. The deviation from the n-paraffins curve increases with the number of rings, the aromatics eluting earlier than the n-paraffins.

The heavy product was prepared for analysis by three different methods:

- solubilization in THF followed by filtration through the syringe
- solubilization in CS_2 followed by filtration through the syringe
- vacuum distillation followed by injection of the different fractions.

Sample analyses of Run 12 starter solvent and product fractions obtained by vacuum distillation are presented in Figure 5.2. The results are only preliminary and these analyses are not quantitative since response factors have not been obtained as yet. However, a considerable spreading of the product relative to the starter solvent is observed. The cuts are not as well defined as expected, displaying some product elution from the columns both above and below the cut points.

Future work will involve vacuum distillation of our product into very narrow cuts and the use of these cuts as calibration standards. The consistency of retention times and response factors of the various cuts will be tested for different products. Considering the presence of polycyclic aromatic and heteroatom containing compounds, calibration in this manner is expected to be more accurate than the standard method mentioned above.

5.2 DETERMINATION OF MOLECULAR WEIGHT DISTRIBUTION

The determination of molecular weight distribution was performed by Gel permeation chromatography with a model ALC/GPC 24 YC Liquid Chromatograph (Waters Associates, Mass.) equiped with a UV detector and a differential refractometer.

The chromatograms were obtained using two styragel columns in series (500A° and 100A°). The solvent was tetrahydrofuran at a flow rate of 1.2 ml/min.

A 0.5% tetrahydrofuran solution of each sample was passed through a 0.5 μ filter prior to injection.

The Refractive index detector continuously measures the difference in refractive index between the reference mobile phase and the mobile phase containing the sample.

Since the detector supplies an output signal that is related to the concentration or weight of solute in the column effluent, the graph is an estimate of the distribution of mole fraction according to molecular weight. However, because in the separation of small molecules retention is only a rough function of solute molecular weight, the chromatogram obtained cannot be used to calculate absolute molecular weights but only to observe modification in the distribution of molecular weight of different products.

MW determinations were conducted on THF soluble product: oils (cyclohexane soluble), asphaltenes (toluene soluble - cyclohexane insoluble), and preasphaltenes (toluene unsoluble - THF soluble).

Figure 5.3 depicts the molecular weight distributions of the THF soluble product obtained in run 10 and run 11. An increase in the concentration of material with molecular weight over 800 could be observed in both runs as the number of passes increases. These can be correlated with the increase in the content of asphaltene and preasphaltene fraction in the product. As expected the use of centrifugation for solvent recovery (Run 11) as opposed to distillation (Run 10) results in a higher concentration of asphaltenes and preasphaltenes in the product.

The heavy product (M4X5) from run 11 was passed through the reactor without catalyst (pass 8) and again with catalyst (pass 9). This treatment did not significantly alter the molecular weight distribution of the THF insoluble or cyclohexane insoluble materials as can be seen from a comparison of the response curves for these two passes.

Typical response curves for the asphaltene and preasphaltene fractions of Run 11 product show a prominent negative refractive index peak which is possibly due to water and/or solvent interferences.

FIGURE 5.1: SIMULATED DISTILLATION CALIBRATION CURVE

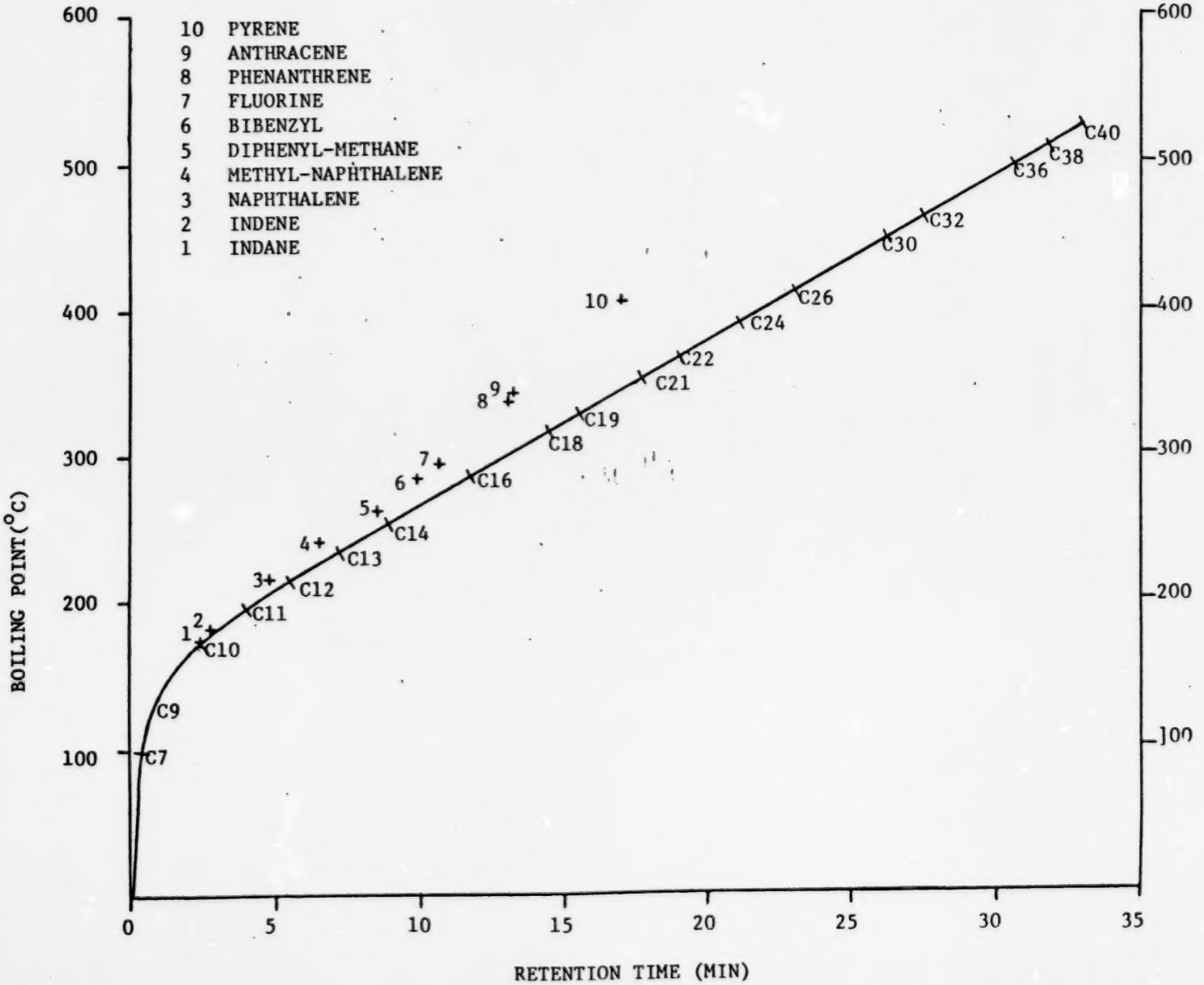


FIGURE 5.2: FID DETECTOR RESPONSE TO RUN 512 STARTER SOLVENT AND HEAVY PRODUCT DISTILLATE CUTS (NOT NORMALIZED)

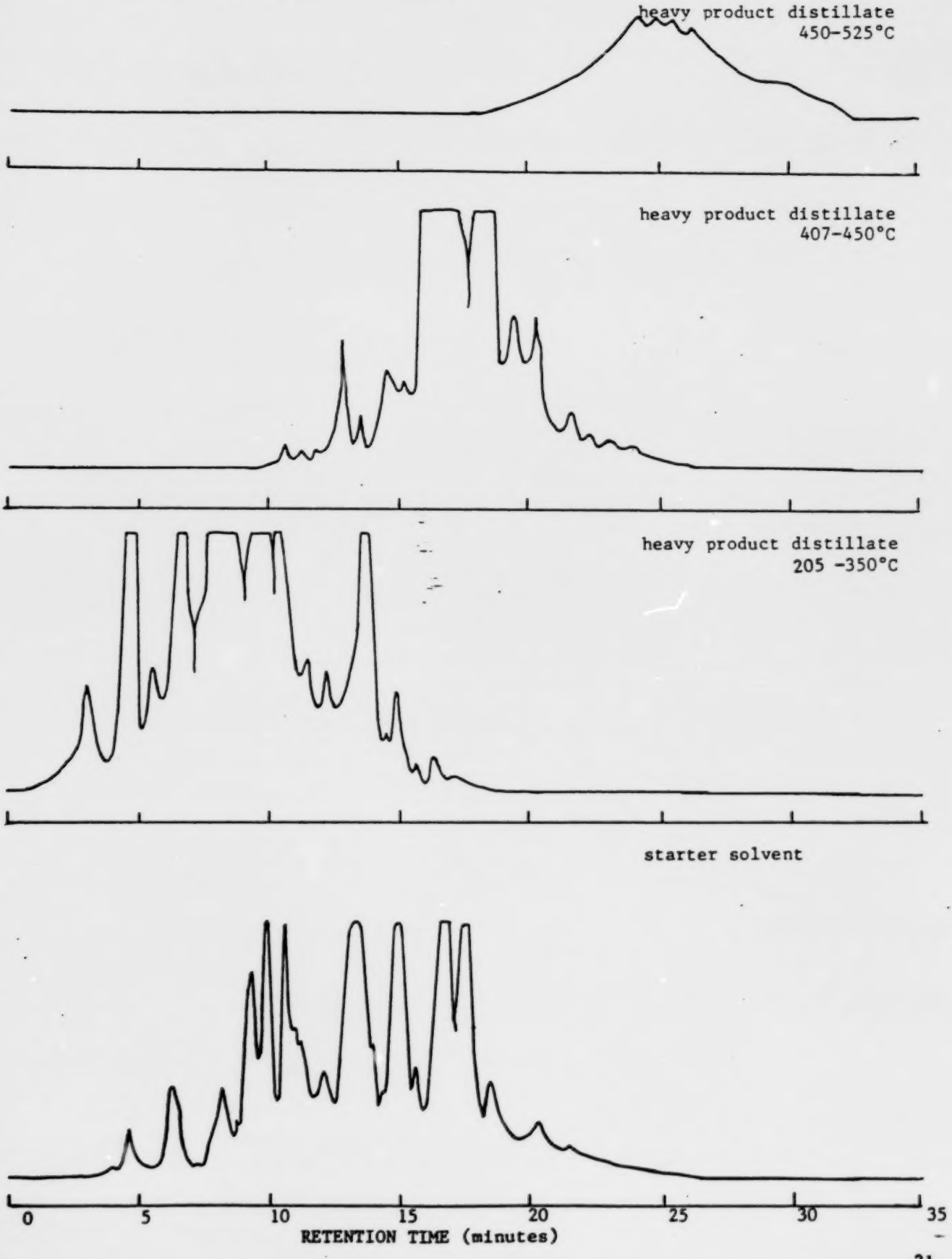
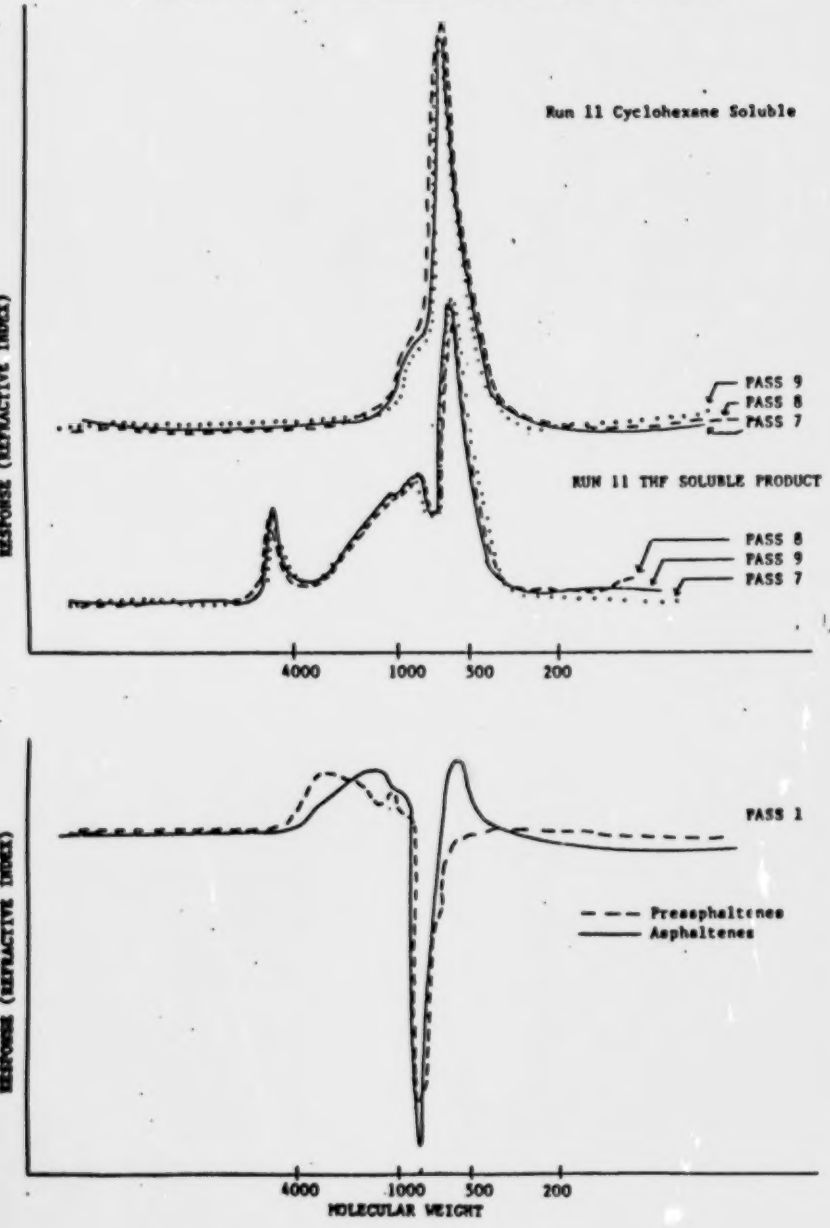
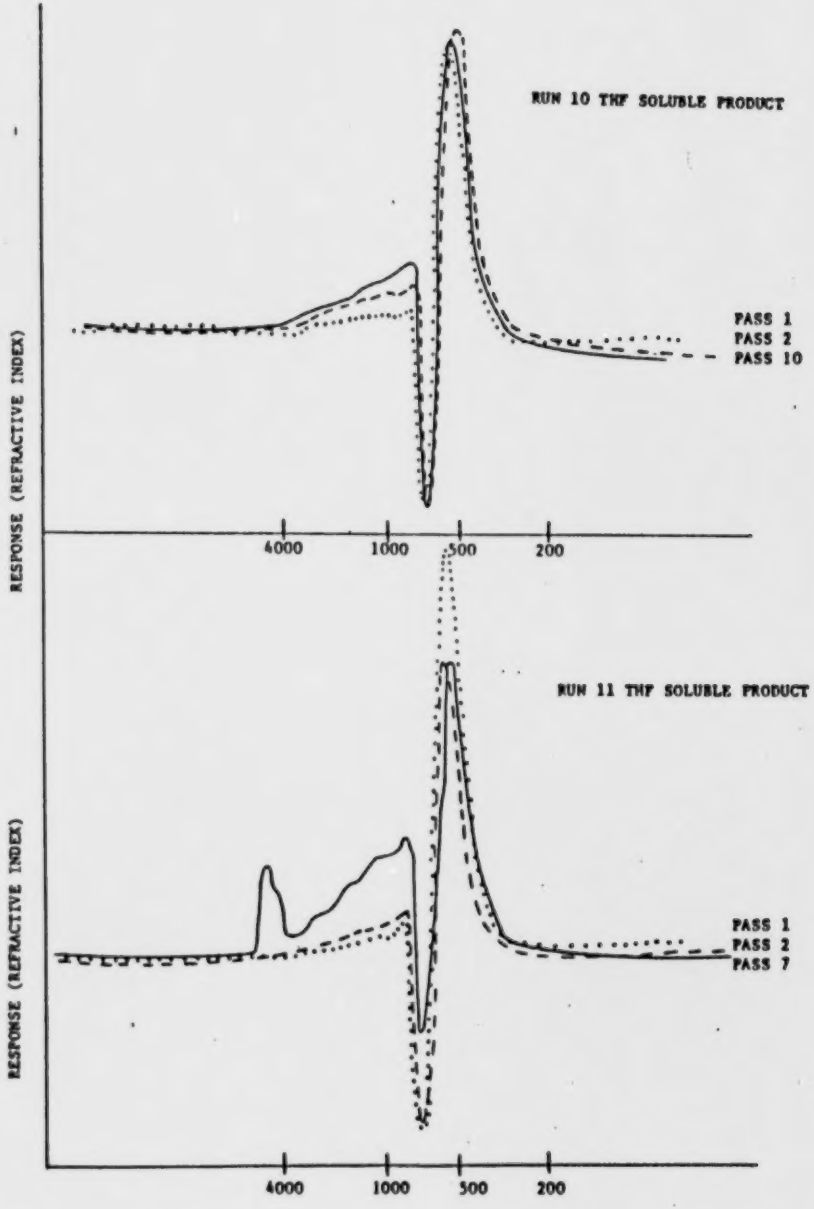


FIGURE 5.3: MOLECULAR WEIGHT DISTRIBUTION OF COAL LIQUEFACTION PRODUCTS OBTAINED BY RPLC - COMPARISON OF RUN 10 AND RUN 11 PRODUCTS





6.0 TESTING PROGRAM

6.1 OBJECTIVES

Three continuous runs were conducted in the recycle mode. The objectives of these runs were as shown in Table 6.1.

RUN 512 was conducted with vacuum distillation of the heavy product as means of removing solids. The main objective was to produce a coal derived product to be used for separate testing of the Sandwell separation process. The solids free products obtained from this separation were mixed into the start-up solvent used for Run 514 .

A secondary objective was to produce hydrogenated coal derived material to be used in solvent quality studies. Commercially available Cobalt-Molybdenum catalyst was ground and slurried in the 250-420°C distillate used as feed.

About 58.0 kg of raw anthracene oil was hydrogenated in the same mode at the end of Run 512.

RUN 513 The objective was to investigate the effect of using hydrogen-carbon monoxide in 1/1 ratio as gas reagent in continuous mode with recycle of the solvent fraction. The goal of completing four passes was not achieved due to mechanical failure.

RUN 514 The objective was to complete four passes in the integrated system using the Sandwell solids separation process for solvent recovery. The start-up solvent was a mixture containing coal derived product. The objective of the run was achieved.

6.2 FEEDSTOCKS

The coal used in the Runs 512-514 was Estevan Lignite of the same shipment used in the testing program undertaken in 1981-82 year. The coal was ground in 1980 and it had been stored under CO_2 atmosphere in drums lined with plastic in order to minimixe the oxidation. In spite of these precautions, the coal aging and oxidation is a possibility that can not be totally disregarded. Each drum of coal was analysed before use for moisture and ash content.

The anthracene oil used in Runs 512 and 513 was part of the 1981 shipment used in the previous years.

The analyses of the coal, start-up solvents and anthracene oil are presented in Table 6.2.

Feed and product analyses were carried out as described in our 1981-1982 report. The addition of a new gas chromatograph allowed for a better analysis of the C_3 and C_4 fractions in the product gas. This chromatograph was also used to analyse the waters physically separated from the M3X3 and M4X6 products.

TABLE 6.1: SUMMARY OF THE CONTINUOUS TESTING PROGRAM

RUN No.	512		513	514
EXP. No.	519	520	522	523
OBJECTIVE	°Produce coal liquid °Reproduce results of Run 10	Produce hydrotreated coal liquid	Test CO/H2 mixture	Simulate integrated operation of SMSS and PDU
FEEDSTOCK				
° coal	Lignite	None	Lignite	Lignite
° starter solvent	Coal derived	Coal derived	AO	Coal derived & AO
° gas composition	100% H2	100% H2	40 CO/60 H2	100% H2
METHOD OF SOLVENT RECOVERY	Vac. Dist.	Vac. Dist.	Vac. Dist.	SMSS
TESTING CONDITIONS				
Reactor temperature (°C)	440°C	400	440	440
Reactor pressure (MPa)	24.2	24.2	24.2	24.2
Solvent/coal (kg/kg)	2.1	-	1.6	2.3
Gas/liquid (L/kg)	174	185	168	181
LHSV (hr-1)	1.08	0.89	0.94	0.91
DURATION OF THE EXP.				
Number of passes	6	1	1	4

TABLE 6.2: ANALYSIS OF COAL AND STARTER SOLVENTS FOR RUNS 512, 513 AND 514

COAL

type: Estevan lignite

moisture (oven) : 22.0%
moisture (xylene): 23.1%
ash (m.f. basis) : 16.7%

SOLVENT COMPOSITION

	ANTHRACENE OIL	RUN 512 STARTER SOLVENT	RUN 514 STARTER SOLVENT
oils	: 95.57%	96.20%	98.23%
asphaltenes	: 4.27%	1.09%	1.71%
preasphaltenes	: 0.13%	0.44%	0.05%
THF insolubles	: 0.03%	2.27%	0.01%

6.3 SOLVENT RECYCLE

In the experiments 519 and 523 the coal slurry feed was prepared by adding fresh coal to a mixture of equal quantities of heavy reactor product containing asaphaltene, pre-asphaltene, ash and unconverted coal, and distillate cut (250 - 420°C). The ratio "solvent/fresh coal" was maintained 2/1 in an attempt to maintain the fresh coal flow into the system constant.

The "solvent" mixture was prepared as shown in Figure 6.1. The heavy product collected from M4X5 over four hours of operation was directed to solids separation, and the solids free product obtained was mixed in a ratio of 1/1 with the heavy product collected in the next three hours. The resulting mixture was then used to slurry fresh coal for the following pass.

A recycle feed prepared in the above manner would be expected to show an increasing ash content since part of the product was recycled directly. An analysis of this case shows that the ash content in the feed will tend towards a constant given by the following equation:

$$\% \text{ ash in slurry feed} = \frac{a \times \frac{cF}{cF-1}}{F} = \frac{ac}{cF-1}$$

Where a = ash content in coal as received (% wt)
F = total feed/weight of dry coal
c = fraction of feed weight recuperated as M4X5 heavy product

Assuming that the condition coal feed/M4X5 recycled/distillate recycled is maintained in the ratio 1/1/1, then F = 3. At the typical conditions of operation in our PDU with lignite as feedstock, the average fraction of feed weight recuperated in M4X5 was consistently 85%.

Ash content for Estevan lignite is 13.0% as received (16.7% m.f. basis). We have,

$$\text{percent ash in feed} = \frac{13.0 \times \frac{2.55}{2.55 - 1}}{3} = 7.1\%$$

A few iterations show that this is the case (to within 0.1%) after only 4 passes.

A total of 120 hours of operation were achieved in three experimental runs with the continuous unit operated in solvent recycle mode. Details of the operation are presented in Appendix 1. Experimental conditions are given in Table 6.1 for Runs 512, 513 and 514. The conditions were similar to those of previous runs with regard to temperatures and pressures and overall gas and slurry feed rates. Differences were in the net coal feed rate and solvent recovery schemes.

6.4 DEFINITIONS AND DATA HANDLING

Data handling procedures and definitions of conversion and yields are basically the same as described in the previous report. Conversion presented as % MAF coal is based on the assumption that the recycled insoluble organic material is inert wereas conversion presented as % I O Feed (insoluble organic feed) is based on the assumption that this recycled material behaves the same as the m.a.f. coal. The following definitions apply:

$$\text{CONVERSION (\% MAF COAL)} = \frac{\text{m.a.f. coal IN} - (\text{THF insol} - \text{IOM IN} - \text{ash IN})}{\text{m.a.f. coal IN}}$$

$$\text{CONVERSION (\% I.O. FEED)} = \frac{\text{m.a.f. coal IN} + \text{IOM IN} - (\text{THF insol} - \text{ash IN})}{\text{m.a.f. coal IN} + \text{IOM IN}}$$

where IOM = insoluble organic matter

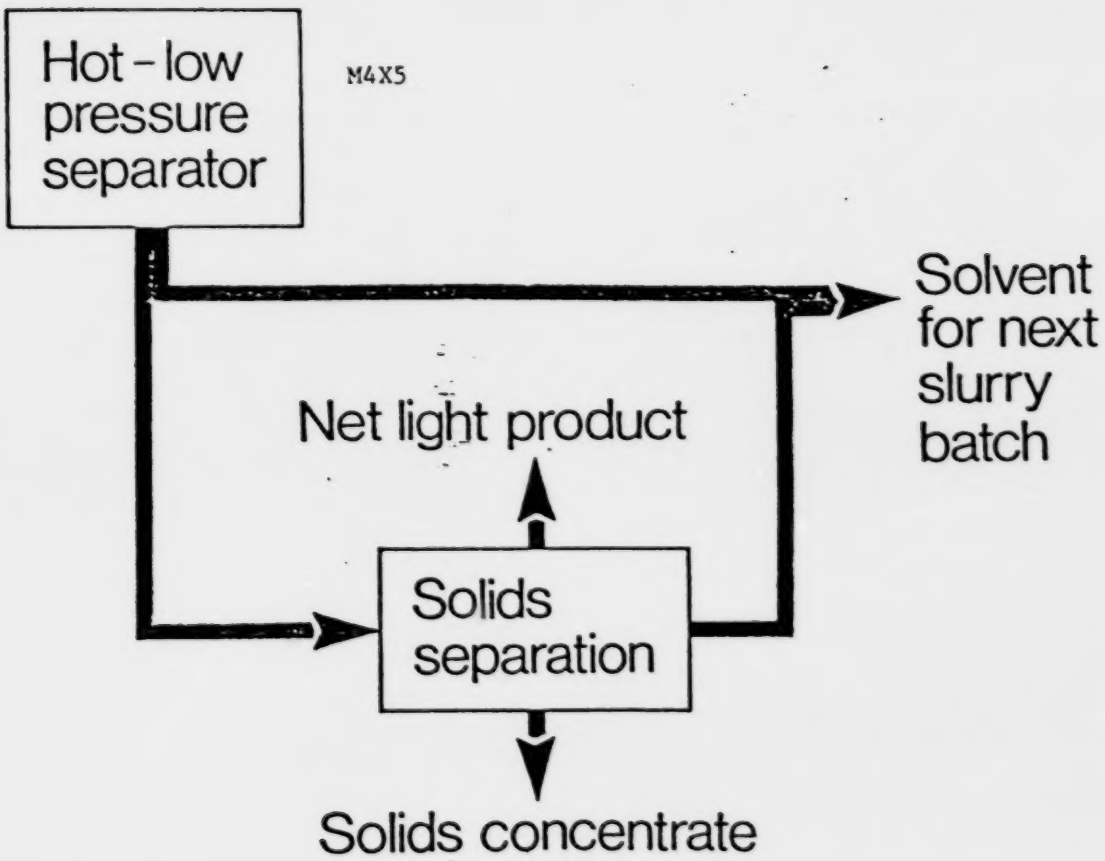
The definition of liquid yield presented below is based on the assumption that the input organic liquids remain unaltered by the reaction. The liquids yields presented in this manner do not quantify absolutely the coal behaviour due to the fact that the input organic liquids may be altered to varying degrees until steady state is reached.

$$\text{LIQUID YIELD} = \frac{\text{THF soluble OUT} - \text{THF soluble IN}}{\text{m.a.f. coal IN}}$$

Hydrogen consumption was calculated using feed and product gas data and not by elemental analysis of the liquids and solids. The following definition applies

$$H_2 \text{ YIELD} = \frac{\text{total H2 in output gas} - \text{total H2 in feed gas}}{\text{m.a.f. coal IN}}$$

FIGURE 6.1: RECYCLE SOLVENT PREPARATION



Data collected over periods of 3 to 7 hours when all operation parameters including feed composition were constant provided basis for material balances, conversion and yields calculation at intervals during each run. The results will be discussed later in this report.

The notion of "pass" was adopted for the purpose of being able to follow the changes in the quality of feed and products and the evolution of the conversion and yields. A pass was defined as the part of the run corresponding to the period when one batch of slurry feed was pumped into the unit. Since each slurry batch was a combination of fractions of product obtained in the previous so called "pass" one can assimilate a "pass" with the passage through the reactor of a specific feed and regard the run as a succession of tests. The average duration of a pass was seven hours corresponding to about seven reactor volumes.

It should be kept in mind that several cycles are required to arrive at a reasonably stable recycle solvent so that "coal behaviour" established by these relatively short runs is an approximation to steady state behaviour. To minimize the number of cycles required to approach steady state, a coal derived starter solvent was used when possible.

In addition to detailed data collected during these mass balance periods, GC analyses of gas effluents and determination of THF insolubles and water content in the feed and heavy product were carried out periodically during the whole time coal slurry was fed. These results were used for a detailed analysis of the whole run which provides a more realistic mass balance and allows for a better appreciation of the coal behaviour since a longer period and larger quantities are involved and small process upsets or material remaining on the walls of vessels become less significant.

Material balances were calculated as a percent weight of the total input as follows:

Gas Stream Calculations

All streams were analysed for the following: H_2, CO, CO_2, CH_4, C_2H_6, C_3H_8, H_2S. The chromatograph data was calculated using external and internal normalization methods and results were checked for consistency. Molecular weight of the gases and percent weight of each component were calculated from the G.C. analysis.

Each stream was regularly monitored for flow and the volumes were plotted against time. A least squares fit of volume vs time was done and total volume per experiment for each stream was calculated. Mass flow rates and total component weight for each stream was calculated from the G.C. and flow data.

Non-gaseous Streams Calculations

The actual feed flow and composition determined on the basis of load cell readings, batch preparation on weights, and average H_2O content in feed (taking into account H_2O losses through evaporation.

The heavy product collection weight and time data was corrected for sampling and spills and a least squared fit was done. The experiment start and end times were then used to calculate total heavy product weight and flow rate. An average % THF insoluble content and a % H_2O analysis of a composite sample were used to calculate the heavy product composition and net weights. Aprioriw was adopted the assumption that ash IN equals ash OUT on the basis that no solids accumulation in the system was observed in any of the tests. Ash analysis was conducted mostly for the solids separation evaluation, not for the material balance purposes.

The light product from M4X6 and M3X3 are separated into water and organic fractions and the weights are plotted versus time to obtain the compositions and weights.

The mass balance was forced to 100% on total input including gases vs total output, and unless there was reason to believe that a certain output weight was in error prorating was done proportionally on all product streams.

6.5 RUN 512 RECYCLE SOLVENT RECOVERED VIA VACUUM DISTILLATION

Run 512 consisted of 5.7 hours of continuous operation during which three experiments (Exp. 519, 520 and 521) were completed.

The experiment 519 was conducted in recycle mode with the objective of producing a large amount of coal derived liquids to be further used in testing of the Sandwell Solids Separation process. The operating conditions selected were similar to Run 10 (see our report for Phase III) i.e. reactor temperature 440°C and reactor pressure 24.2 MPa. The starter solvent used was a combination of coal derived product from previous runs. It was expected that by using coal derived oil as starter solvent, steady state operation would be reached sooner. Six passes were completed in this experiment. A portion of the final product from Experiment 519 was distilled and the 250-425°C cut was used in Experiment 520, the objective of which was to increase the hydrogen content of the oil using a commercial Co-Mo catalyst (HDS 1442) suspended in the liquid phase, in the presence of elemental sulphur and hydrogen at 400°C and 24.1 MPa and a residence time in the reactor of 60 minutes. The third experiment (no. 521) involved catalytic hydrogenation of anthracene oil using the same catalyst at the same conditions.

6.5.1 Experiment 519

The coal used was taken from a freshly opened drum (to minimize the coal oxidation) and the start up solvent was prepared from six parts distillate (the 250°C-420°C cut from previous year's runs No. 8, 10, 11 and 5) and one part heavy product (M4X5) from the last pass of Run 11. The composition of the solvent was shown in Table 6.2.. Pure hydrogen was used once through.

The duration of this run was 37 hours during which 86.0 kg of fresh coal (56.5 kg m.a.f. coal) was fed into the system, and six passes (corresponding to six successive batches of slurry feed) were completed.

Operating History

A mechanical problem interfering with the feed load cell readings invalidated the mass balance data for the first three passes of Exp. 519. Also a total failure of the heavy product let down valve caused a blow down and interruption of the pass 5 for about 80 minutes. Following the blowdown the reactor was cooled to 380°C for about 80 minutes while this valve was replaced. The unit was then started up directly on coal slurry and brought back to temperature and pressure. At this time it appeared that zone 1 (reactor inlet) was very slow in heating up and that the inside temperature at the top of this zone could not be increased above 385°C as compared to 440°C in the upper three zones. The run was continued at these conditions and 250 minutes after the blowdown, a second data collection period (for pass #6) was started. For the remainder of the run, only the upper three zones (about 75%) of the reactor were at the desired reaction temperature while the lower zone indicated only 385°C.

With this exception, the operation was quite stable. Repeated seal failure of the let down valves was experienced without interruption of the run. The post run analysis led to the conclusion that the failure of let-down valves was due to errors in the adjustment of the actuator spring and in the packing procedure. Excessive tightening of the packing caused slow operation of the valve resulting in rapid errosion of valve seals and stems. The design of the PDU's letdown module allowed replacement of these valves without having to stop the test.

A detailed run history is attached in the appendix 1.

Overall Mass Balance

The overall measured material balance for the six passes of the experiment was 98.8% of the input with losses due to product not recuperated from spills or entrained via the back pressure regulator into the scrubber. Product distribution on the continuous unit was as follows:

- 5% Flashed off the hot high pressure pot and collected in M3X3
- 11% Flashed off the hot low pressure pot and collected in M4X6
- 84% Heavy product including the solvent, ash and unconverted coal, collected in M4X5. A portion of this product was used for preparation of the recycle solvent directly, and the other portion was distilled under vacuum for recovery of the 250-420°C cut used for solvent recycle.

Figure 6.2 presents a summary mass balance showing all the flows prorated and normalized to 100 units of m.a.f. coal. This type of presentation does not by any means show the yields obtained at these conditions since the figures extend over the whole duration of the experiment and not only the data collecting periods.

The solvent to fresh coal ratio was unchanged for the duration of the experiment and the slurry flow was maintained steady in order to maintain a constant m.a.f. flow to the system.

A total of 125.7 kg of fresh feed including coal, starter solvent and hydrogen was fed into the reactor during a 37 hour period.

Through vacuum distillation of about 4/7 of the heavy product collected in M4X5, the corresponding proportion of ash, unconverted, asphaltene and preasphaltenes were removed with the residue.

The bottle-neck of the unit operation remained the distillation of the heavy product which being designed for batch operation with a three hour cycle, has limited capacity of processing the heavy stream and does not allow for removal of sufficient solids and polar compounds in order to maintain constant feed characteristics.

The by-products column in Figure 6.2 shows that at the end of the run, 42.10 of the total of 68.15 units of net coal liquid product remained tied in the distillation residue. Only 18.65 of the 20.40 unit of ash input were removed through distillation leading to an accumulation of 2.43 units of ash in the recycle (S_2 - replacement solvent). A similar observation can be made regarding asphaltenes and preasphaltenes which in this experiment showed a gradual accumulation in the recycle solvent, over the six passes of 2.04 units of asphaltenes and 8.5 units of preasphaltenes. These compounds being removed in the form of vacuum bottoms together with the ash and unconverted coal, appear as by-products in this configuration of the process.

It is not expected that steady state was achieved during this experiment. The comparative results of two data collecting periods at 10 hours interval will be discussed further in this report.

Detailed Results

Data from two collecting periods corresponding to passes No. 4 and 6 will be discussed in what follows. The duration of each period was 7.5 hours.

Table 6.3 presents a summary of conditions, mass balance and results for these two data collecting periods. A bar graph of the product yields is presented in Figure 6.3. The discussion is limited to reactor products only, the yield data represent gross figures and do not include the solids separation step. With regard to the operating conditions, the only difference between the two passes was that in pass 6 (which occured after the blowdown incident) the temperature at the top of the first zone of the upflow reactor was only 385°C, or 55°C less than in pass 4. One could argue then, that the actual active reactor volume was reduced for this reason by as much as 25% which might explain why in the sixth pass the conversion was lower and deoxygenation products yield as well as gas yield were also lower.

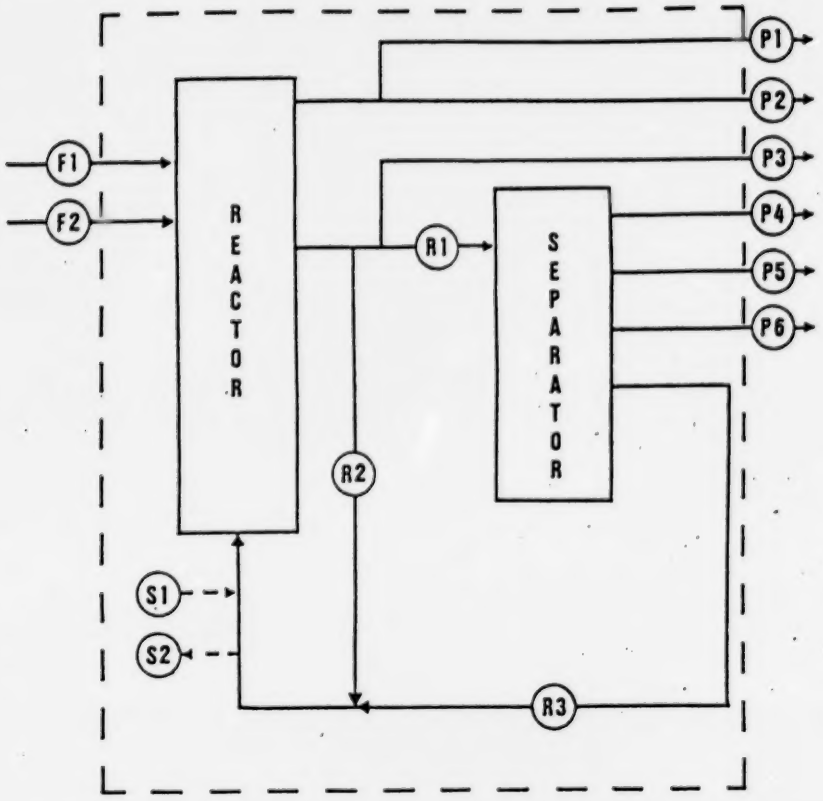
A more probable cause of these findings is the change in the quality of the recycle solvent. Tables 6.4 and 6.5 present a detailed analysis of the feed and product for the two data collecting periods. It appears from the data presented that no marked difference existed between the two feeds with regard to total content of oils, asphaltene, preasphaltenes or non reactive organic insolubles. The distribution of distillation cuts in the products used to prepare the solvent is shown in Figure 6.4. The products from pass 3 (which was used as solvent in pass 4) and pass 5 (which was used as solvent in pass 6) show a difference in the distribution of lighter fractions, but not in that of heavier fractions.

The possible explanation of this apparently odd behaviour of two solvents which are very similar in the distribution of the heavier fractions, which are supposed to be better solvents for coal, might be that the cyclic aliphatic component which is present in the solvent in the beginning of the run, diminished in favour of the atomatic component, as a consequence of donating hydrogen to the coal and coal products. It is then conceivable that would the solvent be subjected to a hydrogenation step before recycle into the system, its quality would stabilize and a steady state would be achieved.

These findings are in agreement with the results published by researchers from Conoco (5,6) in their study of solvent quality for two phase liquefaction.

It is interesting to note though that in spite of the difference in the conversion levels between the two data collecting periods, the gross liquid yield is practically unchanged, while the gas yield and the deoxygenation product yields (H_2O, CO, CO_2) are higher in pass 4 where the reactor temperature was 440°C in all zones. This is accordingly reflected in the higher hydrogen consumption in pass 4.

FIGURE 6.2: RUN 512 EXPERIMENT 519 PROCESS FLOWSHEET AND SUMMARY MASS BALANCE



	NORMALIZED FEED STREAMS			NORMALIZED OUTPUT STREAMS						
COMPONENT	F1	F2	TOTAL FEED	P1	P2	P3	P4	P5	P6	TOTAL OUTPUT
H2	6.70		6.70	3.36						3.36
MAF coal		100.00	100.00							
C1-C4				8.15						6.46
OIL					3.66	5.88	16.51		3.49	29.53
ASPH.									19.05	19.05
PRE-ASPH.									19.57	19.57
H2S				0.16						0.16
ASH		20.40	20.40						18.65	18.65
IO MATTER									11.96	11.96
CO2+CO				8.60						8.60
H2O		21.11	21.11		7.64	20.76	0.77			29.17
TOTAL	6.70	141.51	148.21	20.27	11.30	26.64	17.28	0	72.72	148.21

	NORMALIZED BATCH WEIGHT OF INITIAL AND REPLACEMENT SOLVENTS				NORMALIZED INTERNAL RECYLE STREAMS				RECOVERED BY PRODUCT	BY PRODUCTS
COMPONENT	S1 INITIAL	S2 REPLACE-MENT			R1	R2	R3		STREAMS	STREAM
H2										3.36
MAF coal										
C1-C4										8.15
OIL	70.47	57.24			158.98	119.68	132.67		26.05	3.48
ASPH.	0.79	5.94			16.48	12.42				19.05
PRE-ASPH.	0.31	5.71			15.83	11.91				19.57
H2S										0.16
ASH	0.68	2.43			18.65	13.89				18.65
IO MATTER	0.70	1.49			11.96	8.52				11.96
CO2+CO										8.60
H2O		0.14			0.77	0.61				29.17
TOTAL	72.95	72.95			222.67	167.03	132.67		26.05	122.16

FIGURE 6.2: Continued

LEGEND:

REACTOR = primary liquefaction unit - include slurry preparation, preheating, reactor and product collection

SEPARATION = include the solids separation and solvent recovery step

P_1 = total gas product - as measured off M3X3, M4X6, M5X1 and M6X1

P_2 = light product condensed off the gas stream, as collected from M3X3 high pressure, cold knock off pot

P_3 = light product flashed off the hot low pressure receiver, as collected from M4X6 atmospheric, cold pot

P_4 = distillate product

P_5 = asphaltene - preasphaltene concentrate

P_6 = ash concentrate

R_1 = heavy product containing solids, feedstock for the solids separation

R_2 = heavy product containing solids, recycled into slurry preparation

R_3 = solids free solvent component recycled into slurry preparation

S_1 = starter solvent used in the first pass

S_2 = starter solvent available at the end of the experiment for use in the next run

NOTE: Feed, product and recycle flows and batch initial and replacement (S_1 & S_2) solvents were prorated and normalized to 100 units of m.a.f. coal over total experiment period.

Figure 6.3:

RUN 512 PRODUCT YIELDS (% MAF Coal)
VS PASS NUMBER

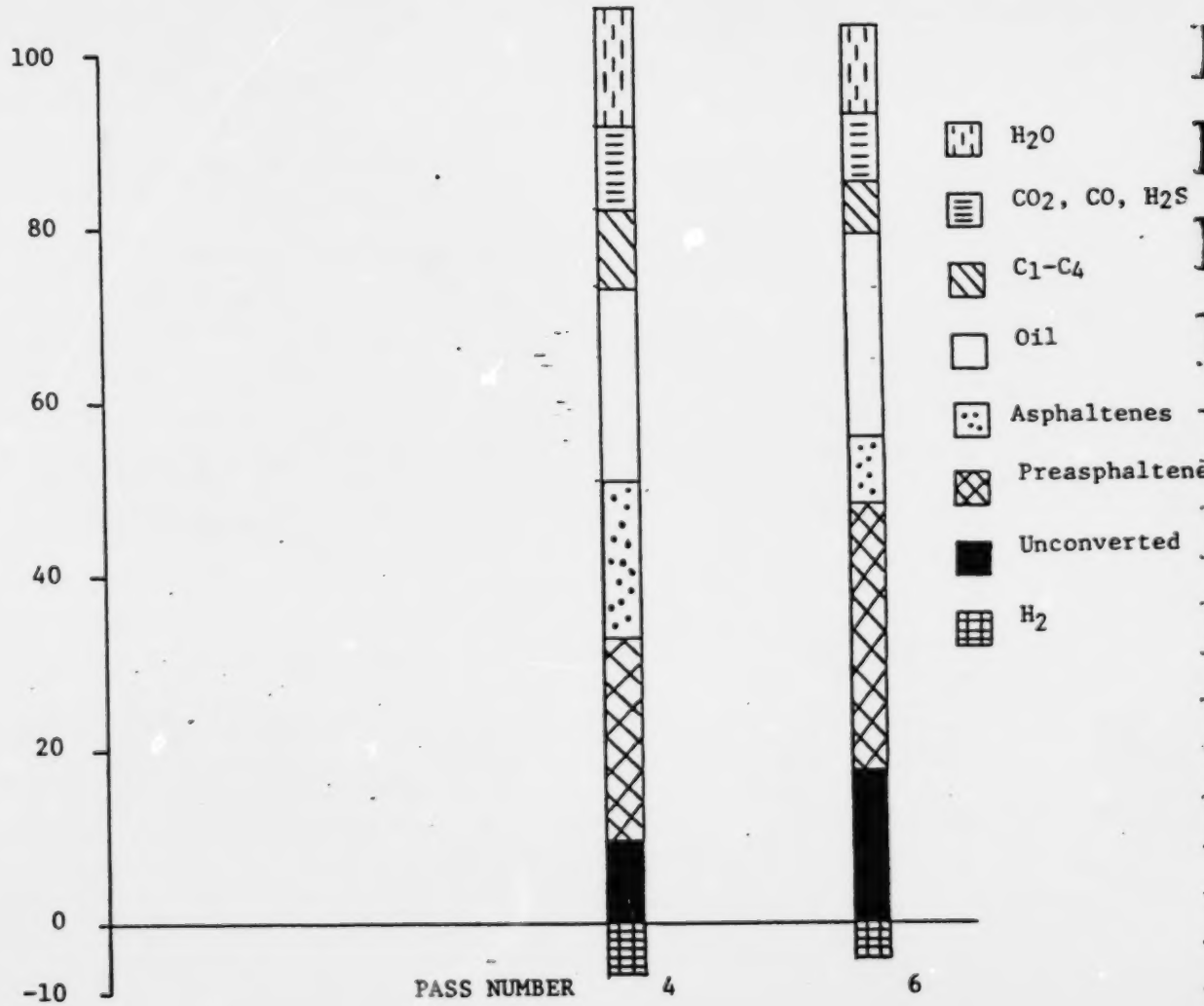


TABLE 6.3: RUN 512 EXPERIMENT 519 OPERATING CONDITIONS AND RESULTS

Pass No.	4		6	
Run Time (hr)	23.5 - 31.0		35 - 42.5	
Preheater Temperature °C				
Slurry	300		300	
Gas	350		350	
Reactor Temperature °C	440		440	
Reactor pressure MPa	24.1		24.1	
LHSV (hr^{-1})	1.08		1.09	
MAF coal flow rate (kg/hr)	1.47		1.52	
Gas/Liquid ratio (L/kg)	172		176	
Material Balance (kg)	**IN**	**OUT (measured)**	**IN**	**OUT (measured)**
H_2O	2.40	3.75	2.44	3.57
Ash	4.31	4.31	3.90	3.90***
IOR**	0.97	2.0	1.23	3.20
MAF coal	10.92	-	11.18	-
Org. Liquid	31.31	37.7	31.22	37.60
H_2	0.773	0.234	0.793	0.430
CO_2	-	0.944	-	0.741
CO	-	0.091	-	0.083
H_2S	-	0.020	-	0.012
CH_4	-	0.296	-	0.206
C_2H_4/C_2H_6	-	0.218	-	0.156
C_3H_8/C_4H_{10}	-	0.393	-	0.298
TOTAL	50.68	49.96	50.76	50.19
Prorated results % maf fresh coal				
Conversion		90.3		82.1
Liquid yield		63.8		61.2
H_2O		12.9		10.4
CO, CO_2		9.6		7.5
H_2S		0.2		0.1
C_1-C_4		8.5		6.0
H_2 consumption		(4.7)		(3.2)

* Only the upper 3 zones were maintained at 440°C, the lower zone temperature was 385°C

** IOR = organic material insoluble in THF (unconverted coal)

*** Ash OUT assumed = ash IN

TABLE 6.4: RUN 512 EXP. 519 COMPOSITION OF FEED AND PRORATED PRODUCT DURING DATA COLLECTION PERIODS*

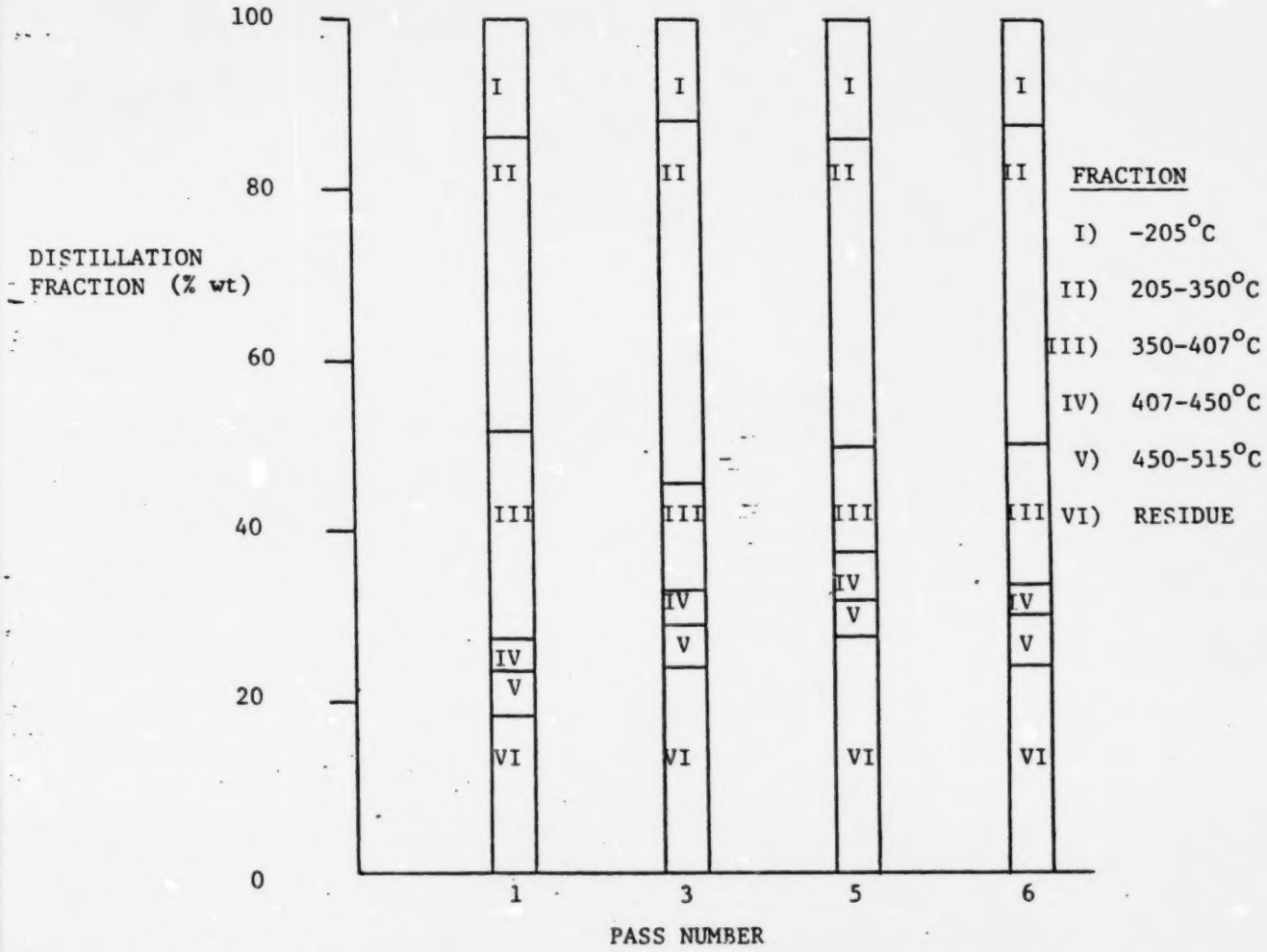
PASS	4			6		
	FEED	PRODUCT	(NET)	FEED	PRODUCT	(NET)
Oil	262.6	285.4	(22.8)	253.8	276.6	(22.8)
Asphaltene	15.2	33.6	(18.4)	13.2	20.8	(7.6)
Pre-asphaltene	8.9	31.7	(22.8)	12.2	42.8	(20.6)
MAF Coal	100.0	0	-	100.0	0	-
IOF Matter	8.9	18.6	(9.7)	11.0	28.9	(17.9)
Ash recycled	19.1	39.5		14.5	34.9	-
fresh	20.4			20.4		
Water	22.0	34.9	(12.9)	21.8	32.3	(10.5)
CO_2, CO	0	9.6	(9.6)	0	7.3	(7.3)
H_2S	0	0.2	(0.2)	0	0.1	(0.1)
C_1-C_4	0	8.5	(8.5)	0	6.4	(6.4)
H_2	7.1	2.2	(-4.9)	7.1	3.9	(-3.2)
Mass Balance		98.6			98.9	

* Figures are prorated and normalized to m.a.f. coal = 100 units

TABLE 6.5: COMPOSITION OF LIQUIDS IN RUN 512

Pass No.	4			6		
	Liquid Composition (% wt)		Net yield	Liquid Composition (% wt)		Net Yield
	Feed	Product	% maf coal	Feed	Product	% maf coal
Oils (hexane solubles)	91.6	81.3	22.8	90.9	81.3	22.8
Asphaltenes (benzene solubles, hexane insolubles)	5.2	9.8	18.4	4.7	6.1	7.6
Preasphaltenes (benzene insolubles, THF solubles)	3.2	8.9	22.8	4.4	12.6	30.6

FIGURE 6.4: RUN 512, EXPERIMENT 519, DISTRIBUTION OF DISTILLATION CUTS IN THE TOTAL LIQUID PRODUCT



An overall material balance and results for the reactor unit (and not solids separation) are shown in Table 6.6. These reflect an average performance for the 37 hours of the experiment. Figure 6.5 shows the evolution of the heavy product composition during this experiment. The increase of THF insolubles and preasphaltenes in the product tends to stabilize after the third pass. The absolute value of conversion average obtained in this experiment is lower than that obtained in Run 10 of last year at similar operating conditions which seem to indicate a loss of reactivity due to coal aging and/or oxidation.

TABLE 6.6: RUN 512, EXPERIMENT 519 OVERALL RESULTS FOR TOTAL EXPERIMENT PERIOD[1)]

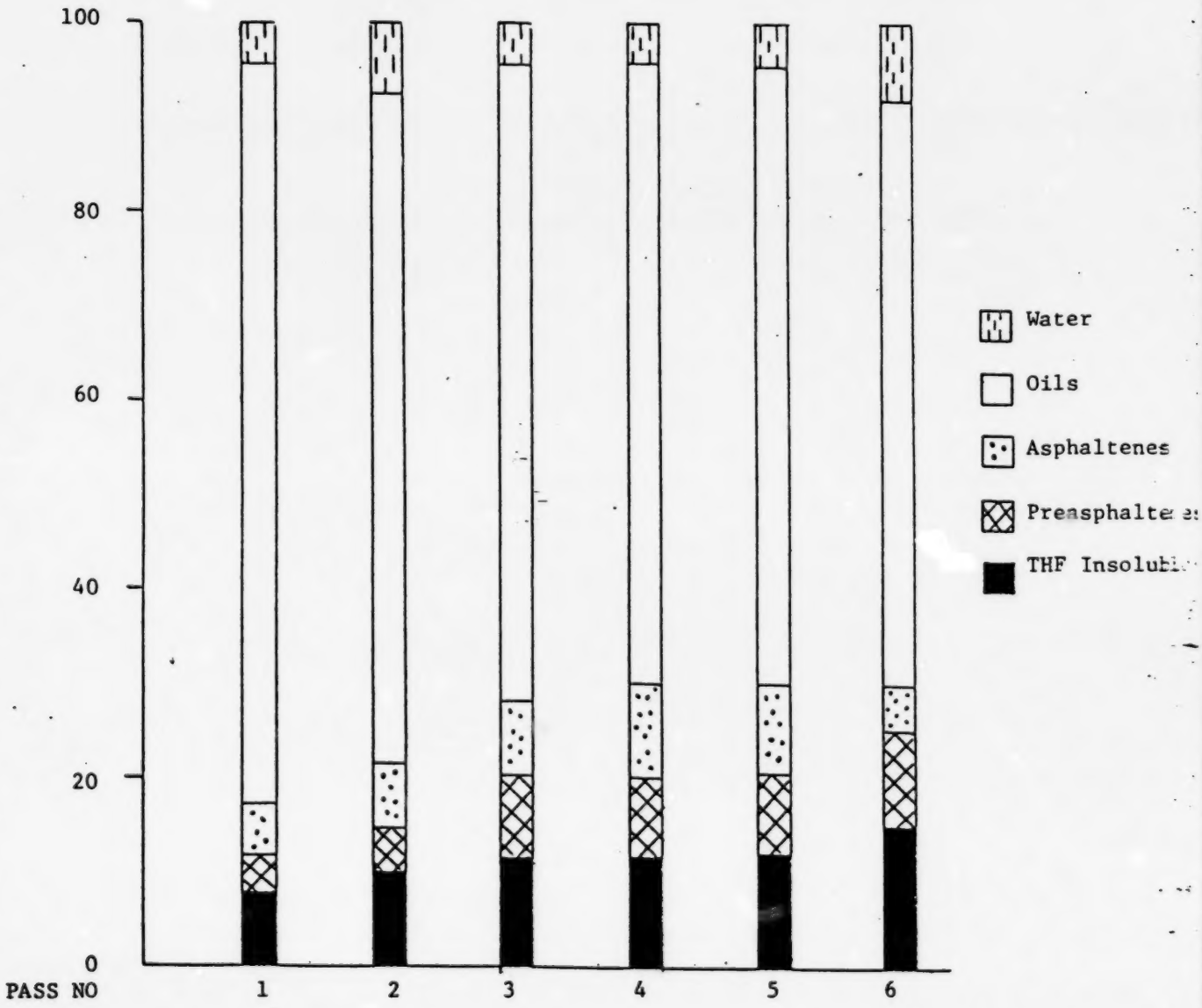
MATERIAL BALANCE	FEED kg	FEED MAF Coal=100	PRODUCT kg	PRODUCT MAF Coal=100
H_2O	12.05	23.32	16.13	28.54
ASH	11.96	21.16	11.96[2)]	21.16
IOR	0.40	0.71	8.68	15.36
MAF Coal	56.52	100.00	0	0
Organic Liquid	40.92	72.40	75.08	132.84
H_2	3.83	6.78	1.89	3.34
CO_2+CO	0	0	4.86	8.60
H_2S	0	0	0.09	0.16
C_1–C_4	0	0	4.60	8.14
TOTAL	125.68	222.36	124.29	219.90

Results % maf coal[3)]

Conversion	85.1
Liquid yield	63.8
H_2O	7.7
CO+CO_2	8.7
H_2S	0.2
C_1–C_4	8.2
H_2	(3.4)

1) Duration of coal feed period 37.1 hours
2) Assumed ash IN = ash OUT
3) Prorated by normalizing products to 100% feed

FIGURE 6.5: RUN 512 EXPERIMENT 519 - EVOLUTION OF HEAVY PRODUCT COMPOSITION WITH THE NUMBER OF PASSES



6.5.2. Experiment 520 - Hydrogenation - Hydrocracking of Solvent Fraction

A slurry composed of 27.1 kg of distillation cut 250-420°C obtained in Experiment 519, 0.27 kg of HDS 1442 catalyst and 0.096 kg sulphur was passed through the reactor at 400°C and 24.1 MPa using hydrogen at a gas/liquid ratio of 185 L/kg. The test lasted about five hours and was plagued with minor mechanical problems such as inconsistent feed flow and poor performance of back pressure regulators, but since the objective was mainly to produce a hydrogenated product, the test was completed in spite of these problems.

The product obtained was distilled and fractions obtained were further used for solids separation tests conducted in the laboratory and then on the PDU in Run 514.

Table 6.7 shows the elemental composition of feed and recovered product distillation cuts. The data appear to indicate that while almost no increase in the hydrogen content occured, some heteroatom removal (nitrogen and oxygen) took place. It appears that hydrocracking was the predominant reaction taking place at the conditions employed. If hydrogenation of the distillate needs to be carried out, it will have to be done in a tricle-bed reactor using a different catalyst probably NiW or NiMo at much less severe conditions, to avoid cracking of the molecules.

TABLE 6.7: RUN 512 EXPERIMENT 520 ANALYSIS OF FEED AND PRODUCT

DISTILLATION RANGE (°C)	FEED wt % distilled	%C	%H	%N	PRODUCT wt % distilled	%C	%H	%N
0-205	0				6.0			
205-350	55.9	90.0	7.78	0.92	59.8	90.3	7.45	0.62
350-407	40.7	91.4	6.23	0.61	29.7	92.1	6.66	0.69
407+	3.4				4.5			

6.6 RUN 513 EFFECT OF CARBON MONOXIDE - HYDROGEN MIXTURE

The objective of this run was to investigate the effect of using a mixture of carbon monoxide and hydrogen as reducing gas in a continuous run with recycle solvent recovered via vacuum distillation at operating conditions similar to Run 512 i.e. reactor temperature of 440°C and pressure of 24.2 MPa, at a slurry flow corresponding to an average retention time in the reactor around 60 minutes. The starter solvent was anthracene oil. The characteristics of coal and solvent are shown in Table 6.2. The gas mixture used contained 60 mol % H_2 and 40 mol % of CO.

Operating History

Only nine hours of operation were achieved before an upset in the gas flow combined with an operating error resulted in coking of the reactor.

A thorough inspection undertaken after the run revealed a 3/4 inch thick ring-shaped coke zone covering the walls of the reactor starting about 0.5 m from the bottom and ending about 0.4 m from the top. It also revealed oily carbon black impurities in the gas line partly blocking the metering orifice on the gas inlet line, which probably was the cause of the eratic gas flow that lead to the reactor coking.

Samples of the coke removed from the reactor were sent to Dr. B. Kybett at the University of Regina laboratory for petrographic analysis.

Discussion of Results

The preliminary results obtained in the first pass completed were interesting enough to warrant a closer look, even though recirculation of the product was not achieved. Very little product was collected in M3X3 and M4X6, a fact that might be due to poor condensation resulting in loss of product into the gas phase. The bulk of the liquid was collected as heavy product in M4X5.

The operating conditions and results are presented in Table 6.8. The material balance was 94.5% of the input with most of the losses probably due to entrainment of water vapours and gases in the scubber water.*

An interesting result is the very high gross liquid yield which at 72.4% of the m.a.f. coal is the highest we obtained to date. An analysis of the composition of this liquid is shown in Table 6.8 and it indicates that relatively high distillable yield can be expected and that the resulting distillates are lower in oxygen and sulphur than the usual lignite derived products.

* a carbon and oxygen balance calculated with the data obtained points toward this assumption.

TABLE 6.8: RUN 513 EXPERIMENT 522 OPERATING CONDITIONS AND RESULTS

Pass No.	1
Run Time (hr)	3.1 - 9.3
Preheater Temperature °C	
Slurry	295
Gas	350
Reactor Temperature °C	440
Reactor pressure MPa	24.2
LHSV (hr^{-1})	0.94
MAF coal flow rate (kg/hr)	1.48
Gas/Liquid ratio (L/kg)	168*

Material Balance (kg)	IN kg	IN (MAF Coal = 100)	OUT (measured) kg	OUT (measured) (MAF Coal = 100)
H_2O	2.36	25.62	0.81	8.79
Ash	1.88	20.41	1.88	20.41
IOR**	0	0	1.09	11.83
MAF coal	9.21	100.00	0	0
Org. Liquid	23.03	250.05	29.70	322.48
H_2	0.360	3.91	0.183	1.99
CO_2	-	-	2.300	24.97
CO	3.456	37.62	1.594	17.31
H_2S	-	-	0.026	0.28
CH_4	-	-	0.194	2.11
C_2H_4/C_2H_6	-	-	0.144	1.56
C_3H_8	-	-	0.334	3.63
TOTAL	40.31	437.68	38.26	415.42

Results % maf fresh coal	
Conversion	88.2
Liquid yield	72.4
H_2O	(16.8)
CO	(20.3)
CO_2	25.0
H_2S	0.3
C_1-C_4	7.3
H_2 consumption	(1.9)

* Average
** IOR = organic material insoluble in THF (unconverted coal)

TABLE 6.9: COMPOSITION OF ORGANIC LIQUIDS IN RUN 513 EXP. 522

	Liquid Composition (% wt)		[1]
	Feed	Product	as measured % maf coal
Oils (hexane solubles)	92.8	84.8	41.6
Asphaltenes (benzene solubles, hexane insolubles)	6.5	9.0	12.8
Preasphaltenes (benzene insolubles, THF solubles)	0.7	6.2	18.2

Elemental composition of the distillation cut 250 - 420°C of the product

C	90.40
H	6.42
N	1.05
S+O	2.13

This could be due to a reaction of the type

$$CO + (O)coal \longrightarrow CO_2$$

where the carbon monoxide reacts with oxygen ether bonds leaving the aryl groups susceptible to stabilization by reaction with the hydrogen. Such a deoxygenation is observed in batch experiments where dried coal and carbon monoxide show a significantly higher carbon dioxide yield than dried coal and hydrogen under similar reaction conditions.

The shift reaction

$$CO + H_2O \rightleftharpoons CO_2 + H_2$$

was quite important as can be seen from the large CO_2 production and H_2O consumption.

The analysis of the results from this run is not complete at the publication of this report but the preliminary data indicate that use of CO/H_2 mixture in direct liquefaction of Saskatchewan lignite should be investigated further.

6.7 RUN 514 EXP. 523 RECYCLE SOLVENT RECOVERED VIA SANDWELL SEPARATION PROCESS

This run's main objective was the commissioning of the first generation prototype equipment for the Sandwell separation process, and the demonstration of the process concepts in an integrated operation with the PDU.

The PDU reactor conditions were similar to those of Run 512 i.e. reactor temperature 440°C, pressure 24.2 MPa and use of pure hydrogen once through at a rate of 190-220 L/kg slurry.

The run was started with a solvent consisting of 60% solids free product (from Exp. 519 and 522) and 40% anthracene oil, and it was carried out for 31.5 hours, during this time four passes through the PDU were completed and the solids separation sequence was carried out three times.

A total of 40 kg m.a.f. coal was fed into the system.

6.5.1 Operating History

Operation of the reactor unit was smooth except for minor variations in the slurry feed rate. This is reflected in the hourly space velocties calculated for each pass. The only upset occured at the end of the third pass when the hydrogen compressor hydraulic circuit failed. For about two hours, while repair work on the compressor was carried out, the gas feed was supplied from the buffer tank M6X2 and gas flow rate, reactor temperature and pressure were lowered.

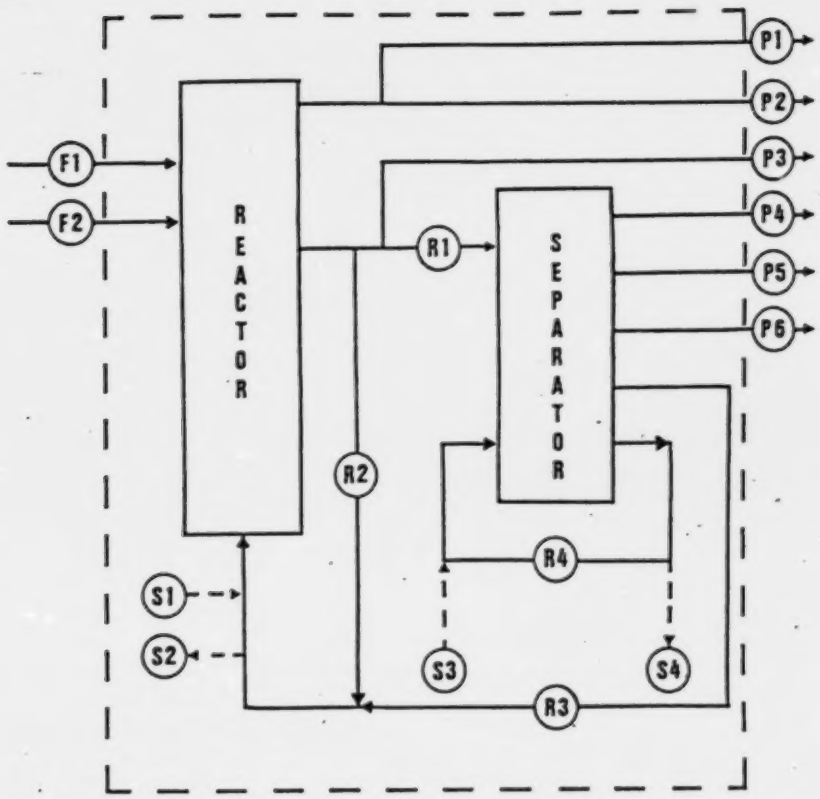
Data collection periods were defined around this upset. A run history is presented in Appendix 1 and the operation of the solids separation unit is discussed in Section 7.

Overall Mass Balance

The measured overall material balance was 95.7% with the losses due mainly to spills or entrainment. Less light products (only 9% of the liquid flow through the reactor) were recovered in the flashed off streams. Most of the reactor slurry stream (92%) was collected as heavy product in M4X5 and it was split into two streams, about 58% being directed to the solids separation and the remaining 42% being recycled as such.

The summary of the material balance for this run is presented in the Figure 6.6 and shows that due to this mode of operation, an accumulation of ash, preasphaltenes and asphaltenes occured in the system. This accumulation was not unexpected and not excessive in view of the fact that the starter solvent for this run was almost 100% distillate material. In our previous work we found it beneficial when the solvent contained some solids and heavy fractions. The accumulation of ash and heavy components in the system can be controlled by a variation of the ratio between the R_1 (portion of heavy slurry stream going to the separation module) and R_2 (portion of heavy slurry stream recycled directly); this deficiency can be overcome.

FIGURE 6.6: RUN 514 EXPERIMENT 523 PROCESS FLOW SHEET AND SUMMARY MASS BALANCE



	NORMALIZED FEED STREAMS			NORMALIZED OUTPUT STREAMS						
COMPONENT	F1	F2	TOTAL FEED	P1	P2	P3	P4	P5	P6	TOTAL OUTPUT
H2	8.19		8.19	4.31						4.31
MAF coal		100.00	100.00							
C1-C4				6.46						6.46
OIL					1.73	4.11	16.52	14.75		37.11
ASPH.								23.31	0.98	24.49
PRE-ASPH.								12.29	0.50	12.79
H2S				0.24						0.24
ASH		20.40	20.40					0.42	14.30	14.73
IO MATTER								1.20	7.41	8.61
CO2+CO				10.14						10.14
H2O		16.00	16.00		0.65	23.89	1.18			25.72
TOTAL	8.19	136.40	144.59	21.15	2.38	28.00	17.70	52.17	23.19	144.59

	NORMALIZED BATCH WEIGHT OF INITIAL AND REPLACEMENT SOLVENTS				NORMALIZED INTERNAL RECYLE STREAMS				RECOVERED BY PRODUCT	PRODUCTS
COMPONENT	S1 INITIAL	S2 REPLACE-MENT	S3 INITIAL	S4 REPLACE-MENT	R1	R2	R3	R4	STREAMS	STREAM
H2										4.31
MAF coal										
C1-C4										6.46
OIL	98.57	83.03	40.89	40.89	182.64	126.31	138.33	115.45	37.11	
ASPH.	4.41	7.41			17.45	17.75			23.51	0.98
PRE-ASPH.	0.12	3.63			8.51	7.86			12.29	0.50
H2S										0.24
ASH		5.68			14.72	13.17			0.42	14.30
IO MATTER	0.03	3.05			6.89	7.31			1.20	7.41
CO2+CO										10.14
H2O		0.33			1.18	0.88				25.72
TOTAL	103.13	103.13	40.89	40.89	231.39	173.28	138.33	115.45	74.53	70.06

FIGURE 6.6: Continued

LEGEND:

REACTOR = primary liquefaction unit - include slurry preparation, preheating, reactor and product collection

SEPARATION = include the solids separation and solvent recovery step

P_1 = total gas product - as measured off M3X3, M4X6, M5X1 and M6X1

P_2 = light product condensed off the gas stream, as collected from M3X3 high pressure, cold knock off pot

P_3 = light product flashed off the hot low pressure receiver, as collected from M4X6 atmospheric, cold pot

P_4 = distillate product

P_5 = asphaltene - preasphaltene concentrate

P_6 = ash concentrate

R_1 = heavy product containing solids, feedstock for the solids separation

R_2 = heavy product containing solids, recycled into slurry preparation

R_3 = solids free solvent component recycled into slurry preparation

S_1 = starter solvent used in the first pass

S_2 = starter solvent available at the end of the experiment for use in the next run

NOTE: Feed, product and recycle flows and batch initial and replacement (S_1 & S_2) solvents were prorated and normalized to 100 units of m.a.f. coal over total experiment period.

The figures presented in the material balance do not represent yields, but they offer a comparison basis to Run 512 where a different concept of solids separation was used. The detailed discussion of conversion and yields evolution during the four data collection periods completed in this run will be presented further in this chapter.

The net improvement of the amount recovered in this experiment is due to obtaining the stream P5 which is very low in solids (we are confident based on our laboratory work that with review of operation procedures and the specially designed equipment, it can be obtained solids free) this stream represents 51% of the coal and is probably amenable to upgrading into distillable products through any one of the available processes (such as LC Fining, Chevron Hydrocracking or CANMET Hydrocracking).

It is not expected that steady state with respect to solvent quality was achieved during this run, but the main objective which was to carry out an integrated operation was achieved successfully and the net product at the end of this run is available for start up of the next run.

Discussion of Reactor Yields and Conversions

Four data collection periods, corresponding to four successive passes through the continuous system, were completed. The conditions and results obtained are shown in Table 6.10. The reactor temperature and pressure were constant and did not change from one period to the next.

The slurry flow varied slightly causing a variation of the liquid hourly space velocity in the range of 0.87 to 0.98. The coal conversion increased in the first recycle pass dramatically from 81.3 to 92% confirming the beneficial effect of returning to the reactor some of the heavy polycyclic compounds and unconverted coal with the recycle solvent.

The same effect is indicated in the gross liquid yield which increased by as much as 16% with the solvent recycle. The somewhat poorer performance in the fourth pass may be due to deterioration of the solvent quality due to the loss of hydrogen donor quality. Data point toward the need of an intermediate hydrogenation step which would eventually ensure steady solvent quality.

Table 6.11 shows the composition of feed and reactor product for each pass.

Table 6.12 presents a detailed distribution of types of components in the liquid product for each pass.

An overall performance of the reactor module for the whole four passes presented in Table 6.13 indicates an average conversion of 90% m.a.f. coal and liquid yield of 67.1 m.a.f. coal.

Through the application of Sandwell Separation Process 97.6% of the liquid product was recovered in solids-free form.

Even though solvent quality steady state was not achieved, solvent balance was maintained for the duration of the run.

TABLE 6.10: RUN 514 EXPERIMENT 523 OPERATING CONDITIONS AND RESULTS

Pass No.	1	2
Run Time (hr)		
Preheater Temperature °C		
Slurry	300	300
Gas	350	350
Reactor Temperature °C	440	440
Reactor pressure MPa	24	24
LHSV (hr^{-1})	0.87	0.95
MAF coal flow rate (kg/hr)	1.07	1.33
Gas/Liquid ratio (L/kg)	203	191

Material Balance (kg)	IN	OUT (measured)	IN	OUT (measured)
H_2O	1.01	2.12	0.46	1.04
Ash	1.46	1.46	1.22	1.22
IOR**	-	1.32	0.30	1.62
MAF coal	7.18	-	4.38	-
Org. Liquid	26.46	29.76	12.96	15.18
H_2	0.661	0.446	0.333	0.148
CO_2	-	0.611	-	0.322
CO	-	0.092	-	0.038
H_2S	-	0.023	-	0.012
CH_4	-	0.154	-	0.065
C_2H_4/C_2H_6	-	0.115	-	0.053
C_3H_8/C_4H_{10}	-	0.184	-	0.094
TOTAL	36.77	36.29	19.65	18.79

Prorated results % maf fresh coal		
Conversion	81.3	92.0
Liquid yield	51.7	67.6
H_2O	15.9	14.4
CO, CO_2	9.9	8.6
H_2S	0.3	0.3
C1-C4	6.4	5.2
H_2 consumption	2.9	4.1

** IOR = organic material insoluble in THF (unconverted coal)

TABLE 6.10: Continued

Pass No.	3		4	
Run Time (hr)				
Preheater Temperature °C				
Slurry	300		300	
Gas	370		370	
Reactor Temperature °C	440		440	
Reactor pressure MPa	24		24	
LHSV (hr^{-1})	0.89		0.98	
MAF coal flow rate (kg/hr)	1.35		1.47	
Gas/Liquid ratio (L/kg)	221		193	
Material Balance (kg)	IN	OUT (measured)	IN	OUT (measured)
H_2O	0.92	1.62	0.87	1.72
Ash	2.11	2.11	2.55	2.55
IOR**	0.36	0.91	0.44	1.26
MAF coal	6.83	-	7.73	-
Org. Liquid	17.70	21.65	20.49	23.70
H_2	0.552	0.231	0.559	0.303
CO_2	-	0.682	-	0.621
CO	-	0.066	-	0.171
H_2S	-	0.015	-	0.012
CH_4	-	0.123	-	0.169
C_2H_4/C_2H_6	-	0.095	-	0.155
C_3H_8/C_4H_{10}	-	0.180	-	0.271
TOTAL	28.47	27.72	32.64	30.93
Prorated results % maf fresh coal				
Conversion	90.9		88.4	
Liquid yield	67.0		60.0	
H_2O	11.0		12.3	
CO, CO_2	11.3		10.9	
H_2S	0.2		0.2	
C_1-C_4	6.0		8.1	
H_2 consumption	4.6		3.1	

** IOR = organic material insoluble in THF (unconverted coal)

TABLE 6.11: RUN 514 COMPOSITION OF FEED AND PRODUCT DURING DATA COLLECTION PERIODS (MAF COAL = 100)

PASS	1		2		3		4	
	FEED	PRODUCT	FEED	PRODUCT	FEED	PRODUCT	FEED	PRODUCT
Oil	351.8	376.9	279.4	311.2	240.0	263.3	240.1	259.5
Asphaltene	17.4	34.4	12.6	34.5	12.9	44.6	17.7	43.9
Pre-asphaltene	0	10.3	3.6	17.5	6.3	18.3	7.2	21.5
MAF Coal	100.0	-	100.0	-	100.0	-	100.0	-
IOR Feed/IOR Residue	0	18.5	6.7	14.8	5.4	14.2	5.5	17.3
Ash	20.5	20.5	27.8	27.8	30.8	30.8	32.9	32.9
Water	13.8	20.7	10.3	25.1	13.3	24.6	11.4	23.6
CO_2, CO	-	9.7	-	8.5	-	11.2	-	11.0
H_2S	-	0.5	-	0.4	-	0.4	-	0
C1-C4	-	6.2	-	4.9	-	5.8	-	4.2
H_2	9.2	6.2	7.6	3.6	7.9	3.3	7.2	4.2
Mass Balance	98.7		95.6		97.4		94.8	

TABLE 6.12: EVOLUTION OF COMPOSITION OF ORGANIC LIQUIDS FOR RUN 514

Pass No.	1			2		
	Liquid Composition (% wt)		Net yield	Liquid Composition (% wt)		Net Yield
	Feed	Product	% maf coal	Feed	Product	% maf coal
Oils (hexane solubles)	95.3	89.4	25.1	94.5	85.7	31.8
Asphaltenes (benzene solubles, hexane insolubles)	4.7	8.2	17.0	4.3	9.5	21.9
Preasphaltenes (benzene insolubles, THF solubles)	0	2.4	10.3	1.2	4.8	13.9

Pass No.	3			4		
	Liquid Composition (% wt)		Net yield	Liquid Composition (% wt)		Net Yield
	Feed	Product	% maf coal	Feed	Product	% maf coal
Oils (hexane solubles)	92.6	80.7	23.3	90.6	79.9	19.4
Asphaltenes (benzene solubles, hexane insolubles)	5.0	13.7	31.7	6.7	13.5	26.2
Preasphaltenes (benzene insolubles, THF solubles)	2.4	5.6	12.0	2.7	6.6	14.3

TABLE 6.13: RUN 514, EXPERIMENT 523 RESULTS FOR TOTAL EXPERIMENT PERIOD

MATERIAL BALANCE[2]	FEED kg	FEED MAF Coal=100	PRODUCT kg	PRODUCT MAF Coal=100
H_2O	6.39	16.00	9.90	24.79
ASH	8.15	20.41	8.15	20.41
IOR	0.01	0.03	3.78	9.46
MAF Coal	39.94	100.00	0	0
Organic Liquid	41.18	103.11	64.78	162.19
H_2	3.270	8.19	1.639	4.104
CO_2+CO	0	0	3.860	9.664
H_2S	0	0	0.095	0.238
C1–C4	0	0	2.459	6.157
TOTAL[1]	98.94	247.72	94.66	237.06

Results % maf coal[3]

Conversion	90.0
Liquid yield	67.1
H_2O	10.0
CO+CO_2	10.1
H_2S	0.3
C1–C4	6.4
H_2	(3.9)

1) Duration of coal feed period 31.5 hours
2) Assumed ash IN = ash OUT
3) Prorated by normalizing products to 100% feed

7.0 PRELIMINARY SIMULATION OF THE SANDWELL SEPARATION PROCESS ON A PDU SCALE

7.1 PROCESS DESCRIPTION

A schematic of the different operations is shown in Figure 7.1.

A certain fraction of reactor product flashed to atmospheric pressure is introduced into a feed vessel. In there it is mixed with a process derived material, denoted here as material 1, and heated to the required temperature for the next processing step.

After attaining the desired temperature, the mixture in the feed vessel is transferred at a controlled rate to a centrifugal device. Nitrogen pressure and a needle valve are used for this. The centrifugal device is a modified laboratory centrifuge. The modified unit has a continuous liquid discharge capability (the overflow) and a relatively large solids hold-up capacity. This allows for the processing of a complete separation cycle in one step. After processing all the material, the solids (centrifuge cake) are manually removed from the centrifuge bowl.

The liquid overflow is processed further in a batch vacuum distillation unit. Here the so called distillates are evaporated off up to the cracking temperature of the material. The non-distillable material is discharged from the bottom of the boiler and is denoted as pitch. Depending on the process conditions this material is either a solid or a very viscous liquid at room temperature. At the boiler operating conditions this material behaves as a liquid and is discharged by gravity drainage. This pitch material is a coal derived liquid product. A fraction of the distillates is utilized in the preparation of the recycle solvent and the rest represents net product.

The centrifuge cake, after manual removal from the centrifuge, is transferred to a custom made vacuum evaporation oven with a large solids handling capacity, high temperature and high vacuum capabilities. In this unit the interstitial distillates are removed from the solids by evaporation. Because of the high distillate content of the interstitial liquid nearly all the coal derived liquids still in contact with the solids are recovered at this stage. Most of the coal derived liquids are separated from the solids in the centrifugation step. The "dried" solids are manually discharged from the oven as a by-product which consists in essence of ash and unconverted coal. The evaporated distillates are condensed and collected for use either as a product or as process solvent.

7.2 DISCUSSION AND PRESENTATION OF PDU SOLIDS SEPARATION EXPERIMENTAL RESULTS

Shown in Figures 7.2, 7.3 and 7.4 are the flows and composition of the process streams for the three separation cycles, carried out in Run 514, Experiment 523. These cycles provided part of the recycle solvent for the four coal liquefaction passes. The other part of the recycle solvent was so called "direct" recycle material obtained from the atmospheric hot slurry receiver (M4X5). The numbers are actual amounts of material processed during the time

Figure 7.1: Flow Schematic of the Simulated PDU Solids Separation Process

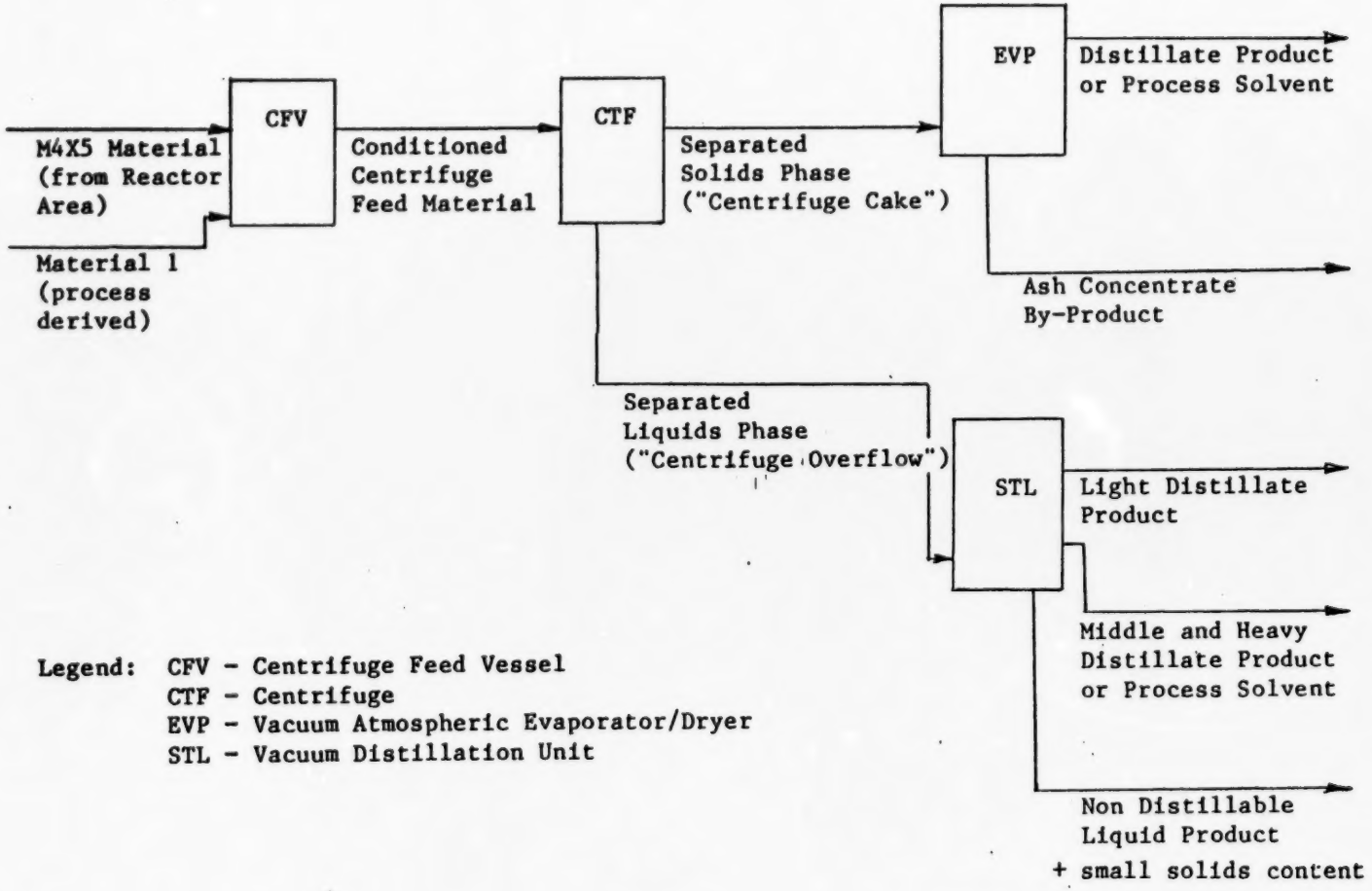
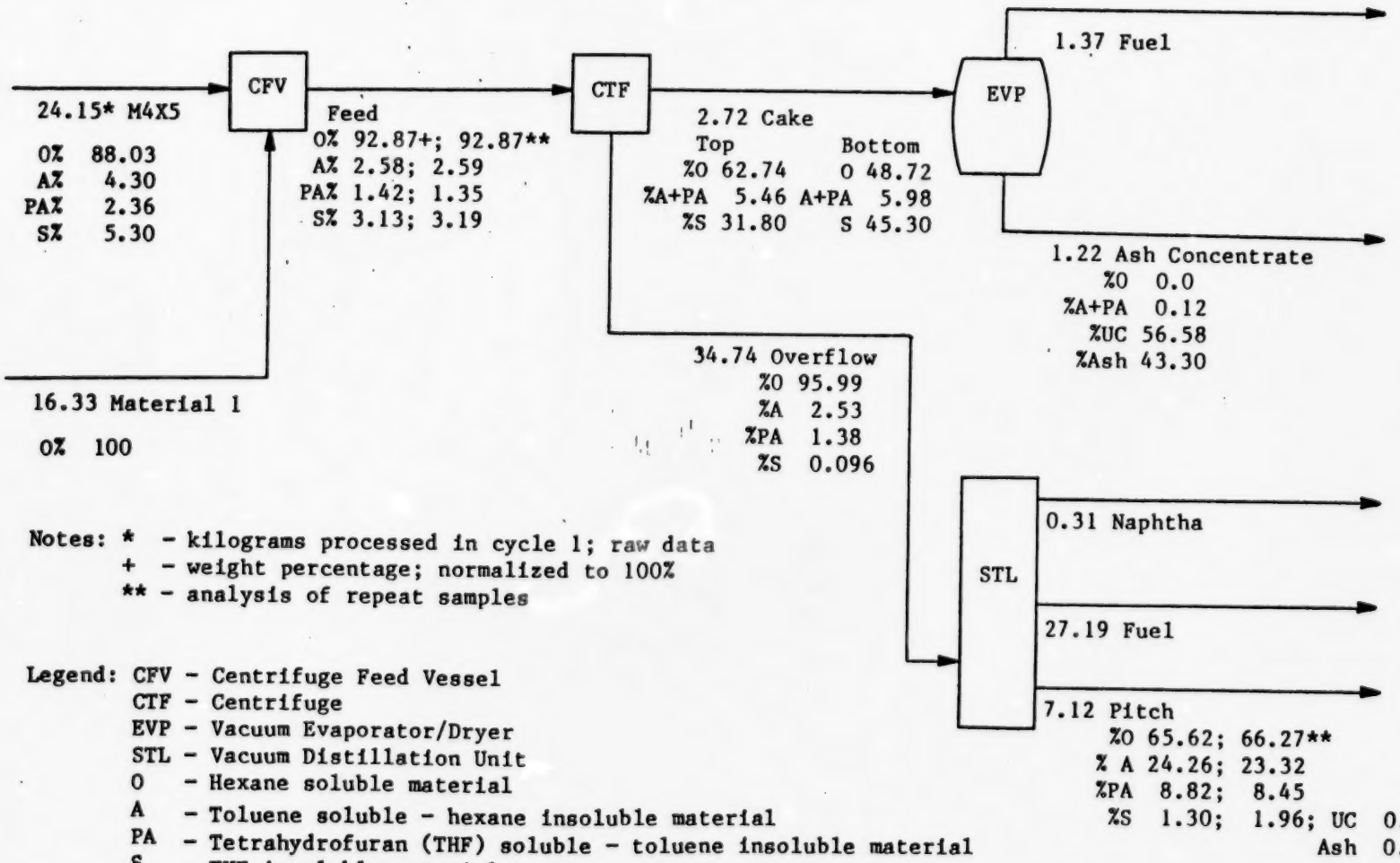


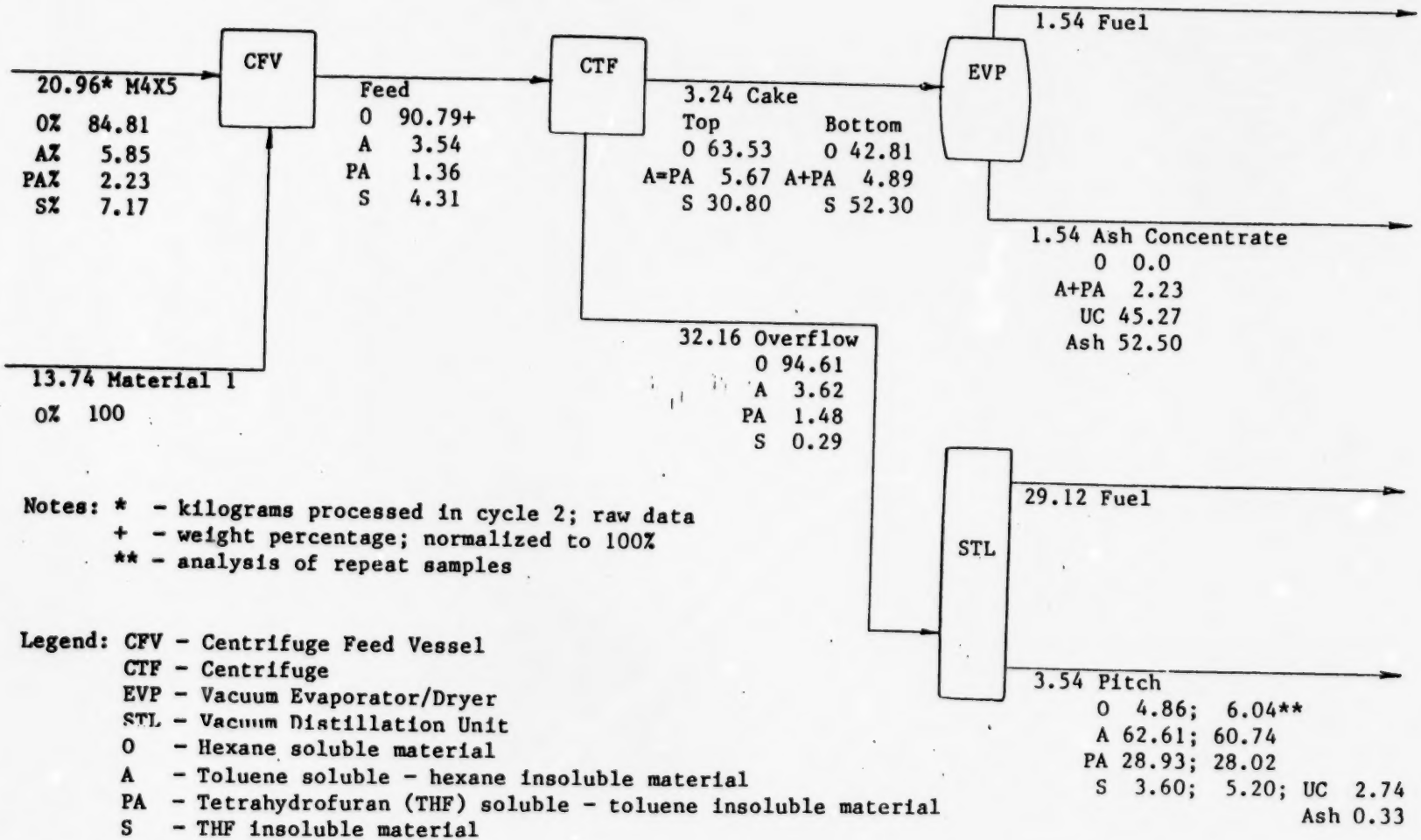
Figure 7.2: Flow Diagram; 1st Separation Cycle; Run 514, Exp. 523



Notes: * - kilograms processed in cycle 1; raw data
+ - weight percentage; normalized to 100%
** - analysis of repeat samples

Legend: CFV - Centrifuge Feed Vessel
CTF - Centrifuge
EVP - Vacuum Evaporator/Dryer
STL - Vacuum Distillation Unit
O - Hexane soluble material
A - Toluene soluble - hexane insoluble material
PA - Tetrahydrofuran (THF) soluble - toluene insoluble material
S - THF insoluble material
UC - Unconverted coal

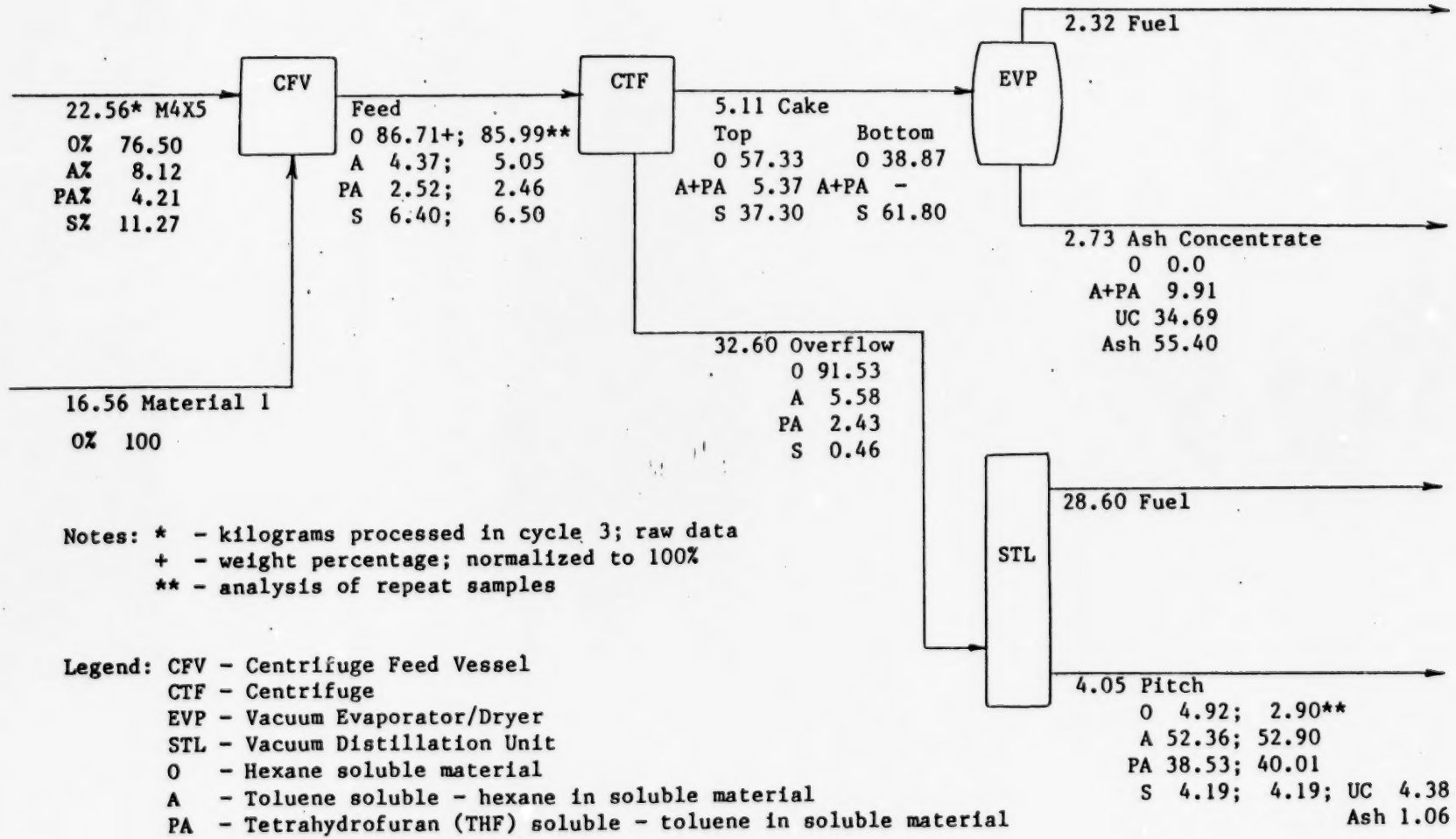
Figure 7.3: Flow Diagram; 2nd Separation Cycle; Run 514, Exp. 523



Notes: * - kilograms processed in cycle 2; raw data
+ - weight percentage; normalized to 100%
** - analysis of repeat samples

Legend: CFV - Centrifuge Feed Vessel
CTF - Centrifuge
EVP - Vacuum Evaporator/Dryer
STL - Vacuum Distillation Unit
O - Hexane soluble material
A - Toluene soluble - hexane insoluble material
PA - Tetrahydrofuran (THF) soluble - toluene insoluble material
S - THF insoluble material
UC - Unconverted coal

Figure 7.4: Flow Diagram; 3rd Separation Cycle; Run 514, Exp. 523



Notes: * - kilograms processed in cycle 3; raw data
+ - weight percentage; normalized to 100%
** - analysis of repeat samples

Legend: CFV - Centrifuge Feed Vessel
CTF - Centrifuge
EVP - Vacuum Evaporator/Dryer
STL - Vacuum Distillation Unit
O - Hexane soluble material
A - Toluene soluble - hexane in soluble material
PA - Tetrahydrofuran (THF) soluble - toluene in soluble material
S - THF insoluble material
UC - Unconverted coal

span of a complete separation cycle. They are actual measurements in kilograms while the compositions are expressed as weight percentages. The differences in the OUT minus IN values are due to: vessels and lines not completely emptying, leaks in pump seals, amounts extracted for samples, and materials collected in cold traps. These are minor amounts and not of relevance to this discussion.

From the composition data of the feed stream to the centrifuge it is seen that the reactor/solids separation system had not reached steady state conditions. That is, the solids, pre-asphaltenes and asphaltene content of the feed increased for each subsequent separation cycle.

The solids content of the centrifuge overflow increases from 0.096% in the first cycle to 0.29% in the second and to 0.46% in the third cycle. This might be a result of the different operating conditions of the centrifuge or it might only be related to the change in the feed composition. This separation step was operated at three very different conditions to find the effect(s) on the solids separation. The second centrifugation was carried out at approx-
imately the same centrifugal force as the first but at more than twice the feed rate (2.1 times). The third one was operated at approximately the same high feed rate as the second but at less than half the centrifugal force (0.4) of the first and second separation. Thus a large G force and a relatively low feed rate appear to result in a more complete solids separation, although the results are not conclusive. This observation is based on very few data points and the results might be compounded by slight changes in type of feed material to the centrifuge.

The amounts of solids removed from the liquid phase in the centrifuge are respectively:

- 1st separation 97.2%
- 2nd separation 93.9%
- 3rd separation 93.8%

These percentages are calculated from liquid stream composition analysis data and weight measurements. Analysis of liquid streams are more consistent than those of solid type materials due to difficulties in obtaining representative samples from a solid mass for analysis. For example, the 97.2% for the 1st separation is calculated as follows:

$$100\% - \frac{0.096 \times 34.74}{\frac{(3.13 + 3.19)}{2} \times 37.46} \times 100\% = 97.2\%$$

The amount of solids not separated at this stage is concentrated in the vacuum bottoms (pitch) fraction for this process configuration. Depending on the effect of these solids in any subsequent upgrading step or in direct product utilization, these concentrations are either acceptable or they need to be reduced. More complex or different process configurations, than the one used here for simulation purposes, should be able to reduce the solids concentration to desired levels.

Also shown in Figures 7.2, 7.3 and 7.4 are the results of ash determinations on the ash concentrate material and the solids fraction of the pitch. Based on these values the amount of ash separated from the liquid product is:

- 1st separation	97.6%
- 2nd separation	98.6%
- 3rd separation	97.2%

This indicates that ash is preferentially separated versus unconverted coal material. Such a result is expected since ash has a larger specific gravity than unconverted coal. Particles with a high ash content should be "easier" to separate than those with a low ash content given that other variables which affect separation are of approximate equal magnitude.

The pitch concentration numbers are analysis of repeat samples. Oil concentrations in these pitch materials varies from about 66% to 5%. This is a function of the distillation "end-point", and provides a certain process flexibility. The 66% material is still a liquid material, although very viscous, at room temperature while the 5% material is a brittle solid at this temperature with a relatively high softening point.

Based on the liquid composition in the feed to the centrifuge and the amounts of distillable and non-distillable liquids recovered and on a material 1 free basis the liquid recoveries are respectively:

- 1st separation	98.6%
- 2nd separation	101.7%
- 3rd separation	98.4%

These are conservative numbers since line and vessel losses, samples and traps amounts are not included in these calculations. In addition, the values are subject to errors which are correlated with separation cycle number. That is, if some of the liquid stays in the system, for instance in separation cycle one and then exits during cycle two, the first liquid recovery value will be too small while that of the second will be too large. Such errors are superimposed on any random errors. This correlation with cycle number explains the 101.7% recovery which is not physically possible, and is the result of this type of error.

8.0 ECONOMIC MODEL

8.1 OBJECTIVES

It is important that process development activities in the field of direct liquefaction be directed into areas having the greatest benefit in reducing the cost to produce coal derived liquids. Any change in the process or an overall process configuration can effect the cost of feedstocks, capital requirement, operating costs and the value of the resultant product mix. Gains in one area may be offset by losses in others thus negating the overall benefit of new approaches.

In addition to the costs factors over which the process developer has some control, there are the external elements which must be endured. These are such factors as interest rates, construction and manufacturing costs, feedstock product revenues. A changing set of conditions could alter the economic advantage of pursuing a given route.

To guide the process development activities of SBRG a simple economic model was developed. The purpose of the model is to:

1) Evaluate the impact on net product revenues due to changes in process conditions or process configuration,
2) Assess the implications of external factors to be able to assess the changes in the economics of coal liquefaction based on structural changes in external factors.

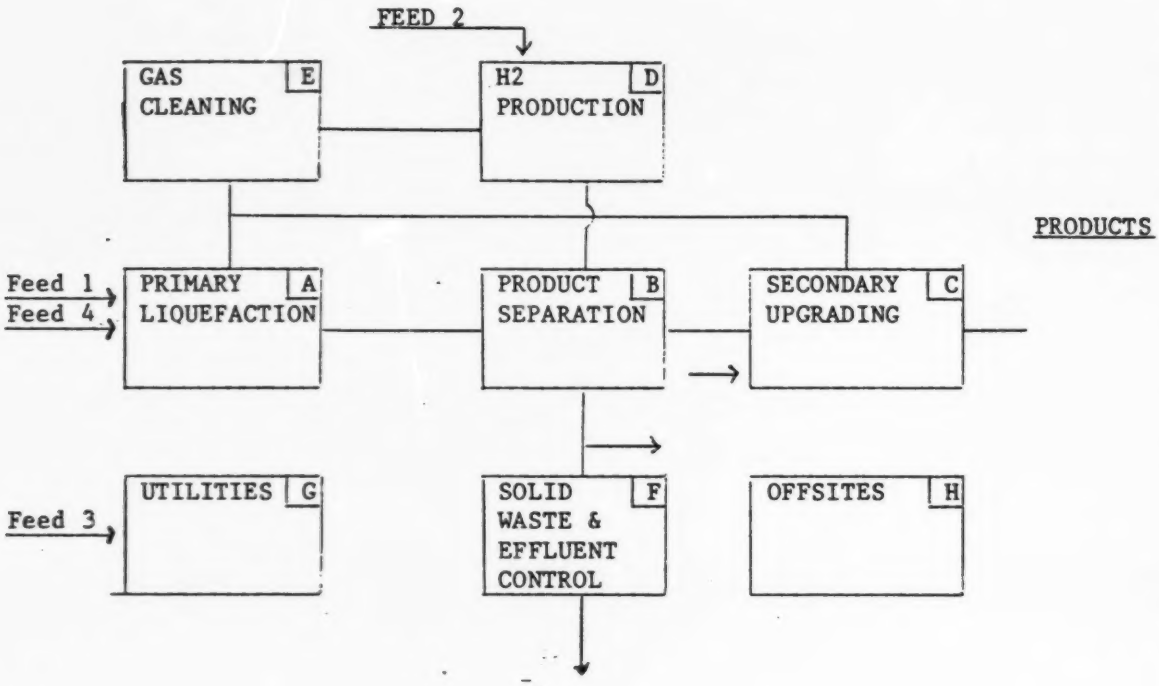
The following describes the model developed and gives some results based on a number of scenarios.

8.2 MODEL REQUIREMENTS

The model to be used for direct liquefaction must be able to have the following features:

- applicable to a broad range of process configurations based on the basic direct liquefaction process
- the case to be modelled must be describable by a mass balance in which the process can be operated in a self-sufficient manner with respect to power and hydrogen requirements
- the capital costs for basic unit operations of the generalized process can be estimated
- the model must be able to handle variations in coal characteristics
- costs are based on commercial scale plants
- the model can handle various feedstock prices and product revenue scenarios
- results are evaluated on the basis of return on investment.

FIGURE 8.1: GENERAL DIRECT LIQUEFACTION UNIT OPERATIONS



8.3 GENERALIZED DIRECT LIQUEFACTION PROCESS

Figure 8. illustrates a general process for direct liquefaction, which can be used to consider the range of feedstocks, products and unit operation to be employed.

8.3.1 Feedstocks

Feed 1) coal

Feed 2) feedstock for production of hydrogen - either, or combination of,
- coal
- process derived methane
- ethane
- LPG
- residual
- purchased natural gas
- purchased hydrogen

Feed 3) feedstock for production of 250 kwh/ton coal of electrical power - either, or a combination of,
- methane
- ethane
- LPG
- coal
- ash concentrate, residuals

Feed 4) catalysts

8.3.2 Unit Operation

The processing plant can be sectioned into units or groups of units performing a given function in the process. These are as follows:

SECTION	NAME	DESCRIPTION	COST FACTORS
A	Primary Liquefaction	coal preparation slurry preparation reactor systems pressure letdown	process conditions, coal throughput
B	Product Separation	liquid/solid separations	process employed, materials handled
C	Secondary Upgrading	non-distillate upgrading as in SRC-1	process used, volume through put
D	Hydrogen Production	generation of H_2 from a variety of feedstocks using optional processes	process used, hydrogen required
E	Emission Control	gas cleaning & scrubbing gas recirculation S recovery NH_3 recovery C1-C4 recovery	process used
F	Solid Waste and Effluent Control	ash disposal effluent treatment	process used
G	Utilities	steam and electrical power generation	process used, power requirements

By this means of sectioning a generalized coal liquefaction plant, a rough indication of capital costs can be determined as process configuration alter the type and sizing of the various process elements.

8.3.3 Products and By-Product Pricing

Net products from the process can be valued according to any pricing scenarios chosen. For a given product slate price, the total revenue can be calculated, or expressed as a value added to the feedstocks or the return on capital invested. Possible products and by-products which could be valued includes:

- methane
- ethane
- LPG
- naphtha
- mid-distillates
- heavy-distillates
- solvent refined coal (ash-free non-distillates)
- residue (vacuum bottom residue)
- S
- NH_3

8.4 ECONOMIC COMPARISONS MODEL

Economic comparison between process alternatives are performed in two ways. The first provides a cost index based on the cost of producing an equivalent barrel of gasoline. The index then gives a relative indication of the marginal differences between various processing cases. It is based on a method used by the Engineering Societies Commission on Energy, Inc. (7).

The cost index is calculated as follows:

$$Cl = \frac{(F + C_A + OP)}{(365\ C_f\ Q_D \leq f_i\ P_i)}$$

where

- Cl = cost index
- F = total annual feedstock and catalyst cost
- C_T = total capitalized cost
- C_A = annual captilalized cost
  $$C_A = r_f C_T$$
- r_f = capital recovery factor dependent on interest rates and amortization period
- OP = annual operating costs
- C_f = annual on-stream capacity factor
- f_i = product value factor of product i
- P_i = amount of product i produced per ton MAF coal
- Q_D = daily F coal feed (MAF basis)

8.5 RESULTS OF USING ECONOMIC MODEL

In order to evaluate the impact of a novel solids separation process such as demonstrated in this study, a comparison was made with the same coal for the processing configurations as shown in Figures 8.2 and 8.3.

Case A is a case where solids separation is performed by vacuum distillation leading to gasification of residues. Electrical power is produced from residuals as well. In case B, solids separation is performed by the Sandwell process, resulting in reforming of C_1-C_4 fraction for hydrogen production and the use of a small amount of additional coal as well as unconverted coal and second stage bottoms for power generation.

Table 8.1 illustrates the approximate capital cost of factors for a commercial scale plant using best estimates based on studies conducted by others (8) and (9) making allowances where it seems appropriate for the lignite coal and the differences in processing conditions. Without further detail cost estimating, well beyond the purpose or the resources of this study, the capital costs cannot be further refined. The total capital cost of around $170,000 per daily ton capacity is in the range of figures usually quoted.

Table 8.2 gives the estimated product distribution and f_i and P_i factors for the two cases. In all cases the products require further upgrading to refined products although this is compensated for by the use of product value factors based on premium gasoline.

Operating costs are also difficult to estimate; however, developers for large plants appear to use 25% of annual capital charges as a first approximation of operating expenses, which in any event will be similar for both cases.

For a comparison of the two cases with the following assumptions, the results are presented in Table 8.3. The net effect according to this model for the case of improved solids separation is a reduction in the cost index of about 27%. This is all due to the greater yield of product from the same amount of coal.

Considering the effect of interest rates the same model shows the significance of a debt financed project at 12% rather than 20%. In this case of the lower interest rate, the cost index is 32% lower. This illustrates the significance of capital charges (either interest rates or construction costs) on the process and the need to find less expensive means of achieving the desired yields.

FIGURE 8.2: DIRECT LIQUEFACTION OF LOW RANK COAL USING VACUUM DISTILLATION FOR PRODUCT RECOVERY

CASE A

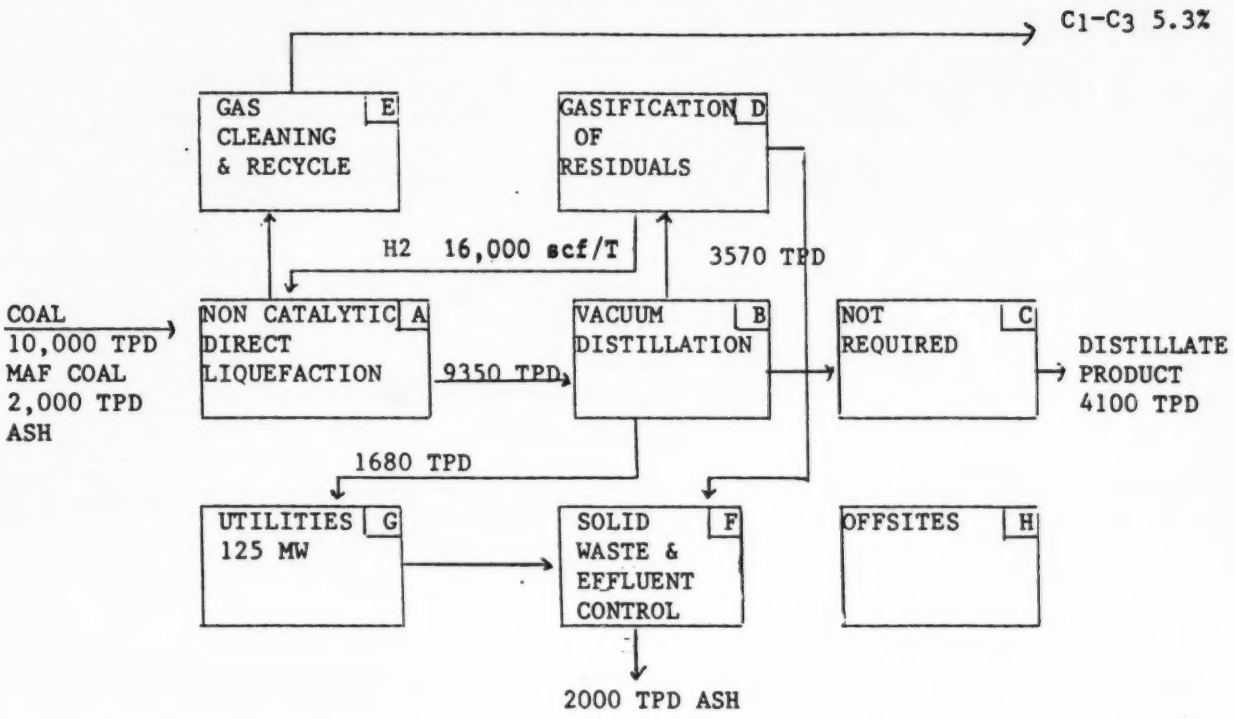


FIGURE 8.3: DIRECT LIQUEFACTION OF LOW RANK COAL USING CONCEPTUAL PROCESS BASED ON SANDWELL SOLIDS SEPARATION PROCESS

CASE B

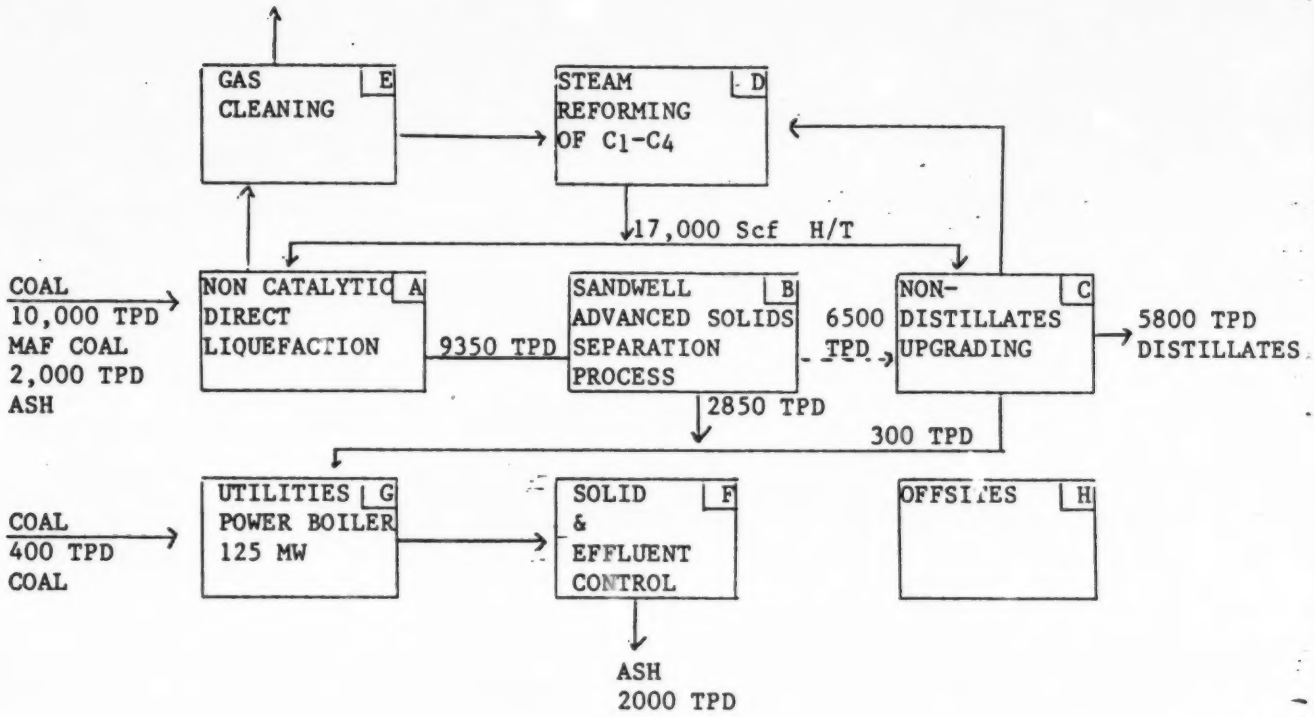


TABLE 8.1: ESTIMATE OF CAPITAL COST SECTION FACTORS

	CASE A	CASE B
	$/daily ton	$/daily ton
A Primary Liquefaction	57,000	57,000
B Product Separation	in A	in A
C Secondary Upgrading	-	12,000
D Hydrogen Production	35,000	21,000
E Emission Control	2,000	2,000
F Solid Waste & Effluent Control	3,000	3,000
G Utilities	11,000	11,000
H Offsites	33,000	33,000
Subtotal	141,000	139,000
Working Capital & Start-up costs	30,000	30,000
	171,000	169,000

TABLE 8.2: ESTIMATE OF CAPITAL COST SECTION FACTORS

PRODUCT	PRODUCT VALUE FACTOR fi	CASE A Pi(Products) bbls/ton	fi Pi	CASE B Pi(Products) bbls/ton	fiPi
LPG	.7	.52	.367		
Naphtha and Light distillates (6.5 bbls/T)	.85	1.04	.884	2.01	1.713
Middle and Heavy distillate (5.7 bbls/T)	.7	1.14	1.798	1.54	1.077
		2.85	2.049	3.55	2.790

TABLE 8.3: EFFECT OF PROCESS AND (20% or 12%) INTEREST RATE ON LIQUID PRODUCT COST INDEX

	PROCESS CASE A		PROCESS CASE B	
Interest Rate	20%	12%	20%	12%
Base Case				
10,000 TPD MAF lignite coal coast $10/T				
C_f = .9				
r_f =	.205	.134	.205	.134
Annual Costs (10^6 $'s)				
F - Annual Feedstocks	36.5	36.5	36.5	36.5
C_A - Annual Capital	350	228.8	346	226.2
Op - Annual Operating Cost	87.5	57.2	86.6	56.6
Total Costs	474.0	322.5	469.1	319.3
Production				
Equivalent bbls of gasoline (10^6)	6.73	6.73	9.16	9.16
Cost Index	70.4	47.9	51.2	34.8

REFERENCES

1. Liquefaction of Low Rank Coals, Report for EMR, Beak Consultants Limited, May 1978.

2. Liquefaction of Low Rank Coals, Report for EMR, Sandwell Beak Research Group, 1980.

3. Production of Oil from Canadian Low Rank Coals Report for EMR, Sandwell Beak Research Group, Jan. 1982.

4. Production of Oil from Canadian Low Rank Coals Report for EMR, Sandwell Beak Research Group, June 1982.

5. Solvent quality effects in SRCI, H Coal and Lummus Two Stage Liquefaction, F.P. Burke, R.A. Winschel presented at "Science and Technology of Synfuels - I", Colorado Springs 1982.

6. Process and Product Oil Characterization in Two Stage Coal Liquefaction F.P. Burke and R.A. Winschel presented at the Eighth Annual EPRI Contractor's Conference on Coal Liquefaction, Palo Alto 1983.

7. Comparison of Coal Liquefaction Process, Final Report on Task 006, The Engineering Societies Commission on Energy, Inc., 1978.

8. Solvent-Refined Coal (SRC-1)-Technology, Product Markets and Economics, Tao, J.C. et al, 3rd International Coal Utilization Exhibition and Conference, Houston, Texas, 1980.

9. Engineering Evaluation of Conceptual Coal Conversion Plant Using the H-Coal Liquefaction Process, AF-1297, Research Report 411-4, Electric Power Research Institute, Palo Alto, California 1979.

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APPENDIX I

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RUN NO. 512 HISTORY

TIME (minutes)		
0	P = 3500 psi H_2 Reactor T = 20°C, heating started	
129	Started pumping warm-up solvent with Milton Roy Stroke set at 35% of full scale Reactor T = 320-270°C	
183	Gamma-ray detector activates right-leg of let down	
260	Reactor temperatures Zone 1 - 440°C 2 - 440°C 3 - 440°C 4 - 435°C	
324	Transfered coal slurry to feed vessel M2x2 THF insolubles in feed 31.2%	(batch 1)
565	Badger Meter valve on right leg not holding pressure - switched to left leg. Replaced seat and stem	
663	THF insolubles in M3X1 9.1%	
696	Transfered second batch to feed vessel THF insolubles in feed 36.0%	(batch 2)
900	Scrubber by-passed for maintenance THF insolubles in M3X1 10.6%	
952	Transfered third batch to feed vessel THF insolubles in feed 33.3%	(batch 3)
1200	Vapor line from M3X1 to M3X2 plugged - it was unplugged by heating the valve	
1277	THF insolubles in M3X1 12.5%	
1395	Transfered fourth batch to feed vessel THF insolubles in feed 36.8%	(batch 4)
1750	THF insolubles in M3X1 15.1%	
1770	Transfered fifth batch to feed vessel THF insolubles in feed 32.9%	(batch 5)
1880	Blowdown - Badger Meter valve on left leg failed at packing Rapture disc at slurry line blew when starting after the blow-down	

1940 restarted pumping coal slurry
Reactor T = 365, 425, 425, 420°C

2069 THF insolubles in M3X1 14.0%

2103 Transfered sixth batch to feed vessel (batch 6)
THF insolubles in feed 34.7%

2150 The Badger Meter valve on the right leg of let-down failed. Switched to left leg

2205 Reactor temperatures
Zone 1 - 383°C
2 - 426°C
3 - 436°C
4 - 439°C

2327 Scrubber by-passed; M5X1 emptied and cleaned - dirt was plugging the NaOH recirculation pump.

2393 Scrubber back in operation

2512 THF insolubles in M3X1 16.6%

2579 Product distillate + catalyst + sulfur
Transfered to feed vessel

2600 Slurry high pressure pump not pumping
Stopped pump, reduced reactor temperature
Reprimed the pump

2611 Start Experiment 520

2927 End of Experiment 520

2951 Hydrogenation of anthracene oil - Experiment 521
Anthracene oil + catalyst + sulfur transfered to feed vessel

3008 H content of M4X5 product 6.8%

3230 Let-down valve leaking - switched legs

3398 Transfered Anthracene oil in M2X2

3405 Started normal shut down

RUN NO. 513 HISTORY

TIME (minutes)	
0	Reactor at temperature, pumping anthracene oil Zone 1 - 400°C 2 - 435°C 3 - 430°C 4 - 420°C Started pumping coal slurry THF insolubles in feed 28.5%
179	Replaced packing in recycle pump
224	M4X5 gas meter malfunctioned - replaced
280	Gas flow very erratic - keeps dropping
525	THF insolubles in M3X1 10.0%
564	Transfered second batch to feed vessel THF insolubles in feed 36.5%
641	Gas preheater outlet temperature increased from 348°C to 368°C - probably due to an undetected drop in gas flow
660	Reactor temperature, Zone 2 started to drop
681	THF insolubles in M3X1 12.5%
730	Temperatures in Zones 2 and 4 hard to control Gas flow keeps dropping
800	THF insolubles in M3X1 15.3%
960	THF insolubles in M3X1 24.0%
975	Transfered batch to feed vessel THF insolubles in feed 42.5%
995	Stopped pumping slurry, cooled reactor
1160	Started feeding pure H_2 Temperature control still difficult. Decided to shutdown and investigate the possibility of reactor coking.
1280	Started feeding anthracene oil
1472	Blowdown Reactor was coked

RUN NO. 514 HISTORY

TIME (minutes)	
0	Started feeding coal slurry - Pass #1 started THF insolubles in feed 25.8% Reactor temperatures Zone 1 - 385°C 2 - 437°C 3 - 440°C 4 - 440°C
357	Centrifugation 1 started
416	Centrifugation 1 finished
400	Valve 2 found closed, compressor not pumping. The system's pressure down to 3200 psi Plug post reactor - condenser. Line before M3X2 was plugged. pressure past reactor 4000 psi B/P regulator 3200 psi Plug was dislodged by heating the line
404	Distillation 5 started
512	Plug at top of condenser - back to normal after unplugging the line
531	M3X3 gas regulator taken apart - full of black oily material
562	THF insolubles in M3X1 7.0%
575	Distillation 5 completed
591	Vacuum Evaporation 1 started
622	Pass #2 started THF insolubles in feed 28.6%
685	Vacuum Evaporation 1 finished
720	Distillation 6 started
747	Distillation 6 completed
758	Slurry flow too high Adjusted the Milton Roy Stroke
802	Vacuum Evaporation 2 started

825 Centrifugation 2 started

847 Distillation 7 started

849 Centrifugation 2 finished

960 THF insolubles in M3X1 10.45%

970 Distillation 7 completed

995 Distillation 8 started

1015 Pass #3 started
THF insolubles in feed 37.0%

1105 Vacuum Evaporation 2 finished

1125 Distillation 8 completed

1197 Line from M3X1 - condenser plugged
Lowered BPR to 2400 & heated the line

1200 -Problem with gas exhaust; outlet pressure 10 psi. Assumed that the bubbler inlet was plugged.
-Bubbler bypassed and gases vented directly

1220 Vent-line unplugged

1272 Centrifugation 3 started

1300 Centrifugation 3 finished

1334 THF insolubles in M3X1 11.5%

1406 THF insolubles in M3X1 12.0%

1425 -Compressor not working
-Lowered reactor temperature

1455 H.P. pump producing erratic flow
-Increased stroke to 38% of the stroke

1460 -Compressor back on stream - it was low on oil
-Increased reactor temperature
-Back pressure increased

1462 Pass #4 started
THF insolubles in feed 37.5%

1479 THF insolubles in M3X1 - 14.9%
-High value due to lower temperature and pressure during the upset between 1425 and 1460

1495 Vacuum Evaporation 3 started

1505 THF insolubles in M3X1 - 14.2%

1562 THF insolubles in M3X1 - 11.6%

1593 -Released 300 psi via #102 to prevent M3X1 - M3X2 line from plugging
-Regulator failed - cannot empty M3X3
-Replaced regulator with manual valve

1625 Vacuum Evaporation 3 finished

1885 THF insolubles in M3X1 - 12.9%

1890 Started normal shut-down procedures